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TOPICAL MEETING ON LASER TECHNIQUES FOR EXTREME
ULTRAVIOLET SPECTROSCOPY 8. (U) OPTICAL SOCIETY OF
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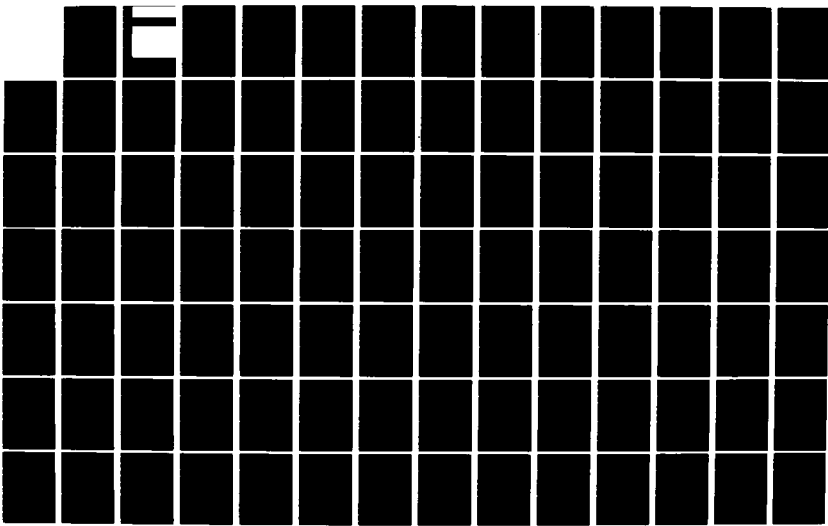
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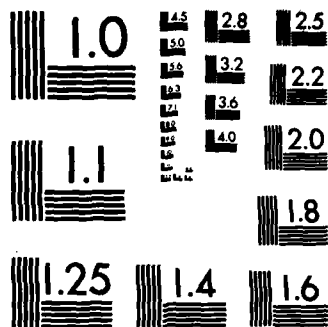
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TOPICAL MEETING ON LASER TECHNIQUES FOR EXTREME ULTRAVIOLET SPECTROSCOPY

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| 19. KEY WORDS (Continue on reverse side if necessary and identify by block number) | | |
| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Twenty years ago, great strides were made in high-resolution spectroscopy in the VUV with the building of large grating spectrographs which provided resolving powers on 23×10^5. Today, with the use of tunable laser sources, resolving powers of 210^5 have been reached in limited wavelength ranges, with 210^5 available over most of the VUV and XUV spectroscopy will be presented, with emphasis on the resolution achieved to date. These results will be illustrated with recently obtained spectra. | | |

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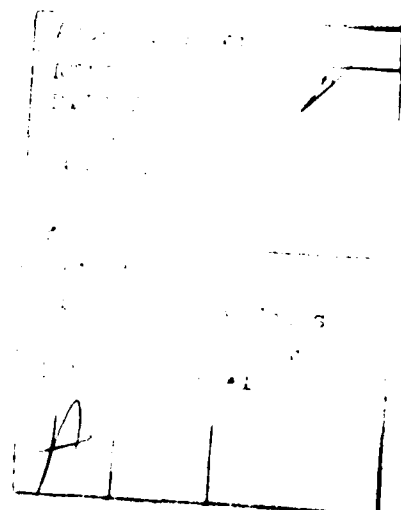
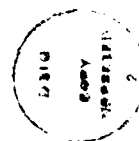
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TOPICAL MEETING ON LASER TECHNIQUES FOR EXTREME ULTRAVIOLET SPECTROSCOPY

**A digest of technical papers presented at the Topical Meeting on Laser Techniques
for Extreme Ultraviolet Spectroscopy, March 8 - 10, 1982, Boulder, Colorado.**

Sponsored by:
Air Force Office of
Scientific Research
and
Optical Society of America



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MONDAY, MARCH 8, 1982

MIDWEST—PACIFIC EXCHANGE

6:20 AM OPENING REMARKS

R. R. Freeman, *Program Co-chairman*

HIGH RESOLUTION SPECTROSCOPY I

R. R. Freeman, *Presider*

8:30 AM MA1 (Invited Paper)

High Resolution Spectroscopy, Thomas McIlrath, *Institute for Physical Science and Technology, University of Maryland, College Park, MD.*

The need for high resolution VUV spectroscopy and the complementarity of grating techniques and laser techniques will be discussed with examples where each is appropriate.

9:00 AM MA2 (Invited Paper)

Tunable, Coherent Sources for High-Resolution VUV and XUV Spectroscopy, B. P. Stoicheff, *Department of Physics, University of Toronto, Canada.*

A brief review of high-resolution VUV spectroscopy using gratings and various light sources will be followed by a description of present laser techniques, with sample spectra.

9:30 AM MA3

Spectroscopic Measurements of Sub-Laser Linewidth Autoionization Rates, W. E. Cooke, S. A. Bhatti, and C. L. Cromer, *Physics Department, University of Southern California, Los Angeles.*

We have simply determined autoionizing rates in cases where the autoionization-induced linewidth is smaller than the linewidth of the laser used to measure the transition's lineshape.

9:45 AM MA4

Laser-Induced Ionization of Na Vapor: A New Mechanism for Producing Na_2^+ , C. Y. Robert Wu, D. L. Judge, J. Roussel*, B. Carre*, P. Breger*, and G. Spiess*, *Department of Earth and Space Sciences Institute and Physics, University of Southern California, Los Angeles, CA.*

The production of Na_2^+ ions by off-resonant laser excitation in the 580-620 nm spectral region mainly results from two-photon resonance three photon ionization in the Na_2 molecule.

*Centre d'Etudes Nucleaires de Saclay, Service de Physique Atomique, Cedex, France.

10:00 AM COFFEE BREAK

MONDAY, MARCH 8, Continued

HIGH RESOLUTION SPECTROSCOPY II

M. Ginter, President

10:15 AM MB1 (Invited Paper)

Optical-Optical Double Resonance Multiphoton Ionization Spectroscopy of NO, M. Seaver, W. Y. Cheung, D. Gauyacq, W. A. Chupka and S. D. Colson, *Sterling Chemistry Laboratory, Yale University, New Haven, CT.*

Double resonance studies using two pulsed lasers were performed on NO yielding highly simplified Rydberg and valence spectra showing effects of internal and external perturbations.

10:45 AM MB2

Frequency Conversion Processes in Hydrogen, Y. Gontier and M. Trahin, *Service de Physique des Atomes et des Surfaces, Centre d'Etudes Nucleaires, Cadex, France.*

Frequency conversion probabilities for resonant and non-resonant multiphoton Raman-like processes taking place above and below the ionization threshold of hydrogen are compared.

11:00 AM MB3 (Invited Paper)

Atomic H and D Concentrations and Velocities Measured With Harmonically Generated Lyman- α (1215 Å) Radiation, R. W. Dreyfuss, *Max-Planck Institut für Plasmaphysik, Garching, West Germany.*

A resonance fluorescence scattering system remotely senses H- and D-atom concentrations in the 10^9 cm⁻³ range in a tokamak. The doppler broadening measures temperatures down to 100°K.

11:30 AM MB4 (Invited Paper)

Laser Induced Energy Transfer in Highly Excited States in Molecules, Jacques Lukasik, Stephen C. Wallace* and William R. Green†, *Laboratoire d'Optique Quantique du Centre National de la Recherche Scientifique Ecole Polytechnique, France.*

The first observation of laser-assisted intermolecular energy transfer between electronic states of carbon monoxide is reported. We also report selective excitation of the b¹ Σ_u^+ valence state (E~13eV) in molecular nitrogen.

*Department of Chemistry and Physics, University of Toronto, Toronto, Canada.

†Watkins-Johnson Company, Palo Alto, CA.

12:00 M MB5 (Invited Paper)

An Overview of Lithium Spectroscopy, Carlos Bunge, *Universidad Nacional Autonoma de Mexico, Mexico.*

12:30 PM LUNCH

MONDAY, MARCH 8, Continued

DIRECT GENERATION I

J. Reintjes, President

2:00 PM MC1 (Invited Paper)

Direct Generation of XUV Radiation, Charles K. Rhodes, *Department of Physics, University of Illinois at Chicago Circle, Chicago, IL.*

The use of rare gas halogen ultraviolet laser technology in the direct generation of XUV radiation will be reviewed.

2:30 PM MC2 (Invited Paper)

Anti-Stokes Raman Lasers, J. C. White and D. Henderson, *Bell Laboratories, Holmdel, NJ*

The first observations of non-resonant, anti-Stokes Raman laser emission are reported. Anti-Stokes Raman scattering of 532 nm and 355 nm pump lasers using inverted metastable T1 atoms resulted in stimulated emission at 376 nm and 278 nm, respectively. Also, a tunable 178 nm anti-Stokes laser using metastable I atoms has been constructed.

3:00 PM MC3 (Invited Paper)

Recombination Lasers in the Vacuum Ultraviolet, W. T. Silfvast and O. R. Wood, II, *Bell Laboratories, Holmdel, NJ.*

Recent observations of new recombination lasers in expanding plasmas include the 4f-3d transition in Li I. Isoelectronic scaling of this sequence suggests a possible 116.8 nm laser in C IV.

3:30 PM REFRESHMENT BREAK

MONDAY, MARCH 8, Continued

DIRECT GENERATION II

R. Gajewski, *President*

4:00 PM MD1 (Invited Paper)

Anti-Stokes Scattering as an XUV Radiation Source, S. E. Harris, R. W. Falcone, J. F. Young, K. D. Pedrotti, Joshua E. Rothenberg, and J. R. Willison, *Edward L. Ginzton Laboratory, Stanford University, Stanford, CA.*

We describe the use of anti-Stokes scattering as a radiation source for high resolution spectroscopy in the 100 Å to 1000 Å spectral region, and as a flashlamp for the construction of XUV lasers.

4:30 PM MD2 (Invited Paper)

Photo-Autoionization Pumped Ba Ion Laser, J. Bokor, R. R. Freeman*, and W. E. Cooke†, *Bell Laboratories, Holmdel, NJ.*

A laser has been demonstrated in Ba⁺ on the Ba⁺ (6p_{1/2}) – (6s_{1/2}) transition following optical excitation of the 6p_{3/2} 12p autoionization state in neutral Barium.

*Bell Laboratories, Murray Hill, NJ.

†Department of Physics, USC, Los Angeles, CA.

5:00 PM MD3 (Invited Paper)

Stimulated Vacuum-ultraviolet Emission From Rare Gas Crystals, N. Schwentner, O. Dössel and H. Nahme, *Institut für Experimentalphysik, Kiel, West Germany.*

Investigations of Xe, Kr and Ar crystals with respect on applications as laser media yield amplified stimulated emission from Ar crystals at 128 nm.

PHILADELPHIA EXCHANGE & GAZEBO LOUNGE

5:30 - 8:00 PM RECEPTION

MONDAY, MARCH 8, Continued

NEW YORK—AMERICAN EXCHANGE

8:00 PM POSTER SESSION

ME1

Laser-Induced Continuum Structure in Multiphoton Ionization Spectroscopy, P. E. Coleman, J. N. Elgin, P. L. Knight, and K. Burnett*, *Blackett Laboratory, Imperial College, London, United Kingdom.*

We report nonperturbative results on multiphoton transition and upconversion enhancement by dressing photolionization continua using radiative interaction analogous to configuration interaction in autoionization.

*JILA and Department of Physics, University of Colorado, Boulder, CO.

ME2

Two-photon Excitation of Argon, Michael S. Pindzola, *Department of Physics, Auburn University, AL.*

The two photon excitation parameter for the ground state to 3p⁵4p¹S excited state transition in argon is calculated in the multi-configuration Hartree-Fock approximation.

ME3

Direct Observation of Hot Electron Spectra from Laser Illuminated Sodium Vapors, J. L. Legouet, J. L. Picque, F. Wullemier*, J. M. Bizau*, P. Dhez*, P. Koch**, and P. Koch†, *Lab. Aime Cotton, Orsay, France.*

Low intensity CW laser was tuned to D₂ line in sodium. Large numbers of electrons with kinetic energy 4.2 and 5.4 eV were observed.

*Lab. Spectroscopy Atomic et Ion and LURE, Orsay, France.

**Yale University, New Haven, CT.

†National Bureau of Standards, Washington, D.C.

ME4

2 π Electron Spectrometer for Multi-Photon Ionization Studies, P. Kruit, F. H. Read* and M. J. Van der Wiel, *FOM-Institute for Atomic and Molecular Physics, Amsterdam, The Netherlands.*

A novel type time-of-flight electron spectrometer, using magnetic fields, is described. It features full collection over 2 π sr at a resolution of 20 meV.

*Schuster Lab., Dept. of Physics, The University of Manchester, United Kingdom.

ME5

Autoionizing and High-Lying Rydberg States of Lutetium Atoms, C. M. Miller and N. S. Nogar, *Los Alamos National Laboratory, Los Alamos, NM.*

Two color multiphoton ionization has been used to examine the spectroscopy of Lu atoms near the ionization threshold of 5.4 electron volts.

MONDAY MARCH 8, Continued

ME6

High Average Brightness Rare-Gas Halide Laser Technology, Stephen E. Moody, George J. Mullaney, William Grossman, Philip E. Cassady, and Stanley Byron, *Mathematical Sciences Northwest, Inc., Bellevue, WA.*

We describe several high repetition rate (100-1000Hz) discharge pumped rare-gas halide laser system having near diffraction limited divergence, resulting in extremely high average brightness.

ME7

Analysis of VUV and Soft X-ray Lasing Based on Charge-Exchange Mechanisms, J. C. Bellum, W. W. Chow, K. Druhl and M. O. Scully, *Institute for Modern Optics, Department of Physics and Astronomy, University of New Mexico, Albuquerque, NM.*

We analyze charge-exchange mechanisms for long-wavelength lasers in terms of the roles played by the primary charge-transfer and other competing processes.

ME8

Resonantly Enhanced Multiphoton Ionization and Third-Harmonic Generation in Carbon Monoxide Gas, James H. Glowina and Robert K. Sander, *University of California, Los Alamos National Laboratory, Los Alamos, NM.*

Three-photon resonantly enhanced multiphoton ionization and third-harmonic emission using the carbon monoxide A state are detected. Pressure and phase matching effects on the VUV emission are discussed.

ME9

Two-Photon Excitation of the Lowest g-States of Xe₂⁺, W. Gornik, E. Matthias, and D. Schmidt, *Institut für Atom- und Festkörperphysik, Freie Universität Berlin, West Germany.*

Two-photon excitations of molecular g-states belonging to the 5p⁵6s configuration of Xe I were measured with frequency-doubled light of a tunable pulsed dye laser.

ME10

Multiresonant Two-Photon-Absorption-Induced Four Wave Mixing in Crystalline Rare Earth Insulators, R. L. Cone, D. A. Ender, M. S. Otteson, Paula L. Fisher, J. M. Friedman* and H. J. Guggenheim*, *Montana State University, Bozeman, MT.*

Coherent nonlinear generation of $\omega_4 = \omega_1 + \omega_2 + \omega_3$ exhibits strong ω_1 (intermediate) and $\omega_1 + \omega_2$ resonances in Tb(OH)₃ and LiTbF₄, providing a novel method for high resolution measurements of excited configurations of rare earth ions using tunable visible and near ultraviolet lasers.

*Bell Laboratories, Murray Hill, NJ.

MONDAY, MARCH 8, Continued

ME11

Space Resolved Spectra of 2p - 3d Multiplets of Al X and Al XI Emitted from Laser Irradiated Thin Al Foils, M. A. Khan and G. J. Tallents, *Dept. of Physics, University of Petroleum & Minerals, Dhahran, Saudi Arabia.*

We will show the spectra of 2p-3d multiplets of Al XI ($\lambda \approx 52\text{\AA}$) and Al X ($\lambda \approx 55\text{\AA}$) emitted from plasmas produced by the irradiation of 9 μm thick Al foils with a neodymium laser focused to peak intensities $\sim 2 \times 10^{15} \text{ W cm}^{-2}$.

*Laser Physics Laboratory, Department of Engineering Physics, Research School of Physical Sciences, The Australian National University Canberra, Australia.

ME12

Resonant Four-wave Mixing Processes in Xenon, Yun Mui Yiu, Keith Bonin and T. J. McIlrath, *Institute for Physical Science and Technology, University of Maryland, College Park, MD.*

Resonant four-wave mixing processes have been used to generate coherent radiation near 125.9 nm and 101.7 nm in xenon.

ME13

Vacuum UV Laser Induced Fluorescence of the NO Molecule, H. Scheingraber and C. R. Vidal, *Max-Planck-Institut für Extraterrestrische Physik, Garching, West Germany.*

Spectrally resolved vacuum UV fluorescence spectra for state selective excitation of different levels of the NO molecule are presented. J-dependent Franck-Condon factors and branching ratios for different electronic transitions have been measured.

ME14

Transform-Limited Bandwidth Injection Locking of an XeF Laser, Irving J. Bigio and Michael Slatkine, *University of California, Los Alamos National Laboratory, Los Alamos, NM.*

We report transform-limited-bandwidth injection locking of an XeF unstable resonator with an Ar-ion laser at 3511 \AA . Sixty percent of the inhomogeneously broadened XeF output was available for locking.

ME15

Excitation and Ionization of Atomic Hydrogen by Narrow Bandwidth Tunable VUV Laser Radiation, H. Zacharias*, H. Rottke and K.H. Welge, *Fakultät fuer Physik, Universität Bielefeld, West Germany.*

Excitation of an atomic beam of H atoms with narrow bandwidth ($\Delta\nu_{\text{VUV}} \approx 0.02 \text{ cm}^{-1}$) circularly polarized tunable VUV laser light leads to the population of single m_F states in 2p $^2P_{1/2}$. Further excitation with a second laser to high Rydberg states and to ionization will be reported.

*Current address: IBM T. J. Watson Research Center Yorktown Heights, NY.

TUESDAY, MARCH 9

MIDWEST—PACIFIC EXCHANGE

PLASMAS I

T. Lucatorto, *Presider*

8:30 AM TuA1 (Invited Paper)

Review of Upconverted Nd-Glass Laser Plasma Experiments at the Lawrence Livermore National Laboratory, K. R. Manes, *Lawrence Livermore National Laboratory, Livermore, CA.*

Much has been learned about laser heated disk target plasmas which may find application in diverse technologies.

9:00 AM TuA2

Resonantly-Pumped Soft X-ray Lasers Using ICF Drivers, Peter L. Hagelstein, *Lawrence Livermore National Laboratory, Livermore, CA.*

Two-dimensional numerical simulations of resonantly-pumped short wavelength lasers indicate that gains between 10-50 cm^{-1} may be obtained by photo-pumping K-shell transitions in one- and two-electron F and Ne ions.

9:15 AM TuA3 (Invited Paper)

Generation of Vacuum Ultraviolet Radiation by Plasma Nonlinearities, R. L. Carman, *University of California, Los Alamos National Laboratory, Los Alamos, NM.*

Numerical simulations of the relevant plasma Hydrodynamics indicate that ten (19.3 nm) or more harmonics of an ArF laser should be generated at high efficiency in a high irradiance solid target plasma interaction experiment.

9:45 AM TuA4

A Soft X-ray Source Based on the Magnetic Confinement and Compression of a Laser Produced Plasma, E. A. Crawford, A. L. Hoffman, and D. C. Quimby, *Mathematical Sciences Northwest, Inc., Bellevue, WA.*

We have demonstrated high beta magnetic confinement of a laser produced plasma. Efficient production of soft X-rays by compression of these plasmas will be discussed.

10:00 AM COFFEE BREAK

TUESDAY, MARCH 9 Continued

PLASMAS II

R. Madden, *Presider*

10:30 AM TuB1 (Invited Paper)

A Laser Produced Plasma as an Extreme Ultraviolet Continuum Source for Absorption Experiments, E. Jannitti, P. Nicolosi and G. Tondello, *Centro Gas Ionizzati, C. N. R., Università di Padova, Padova, Italy.*

The continuum emission from plasmas produced by laser interaction with plane targets has been absolutely measured in the XUV. Application as a source for an absorption experiment of highly ionized species is described.

11:00 AM TuB2 (Invited Paper)

Optical Techniques for Use with Soft X-ray and XUV Radiation, D. T. Attwood and N. M. Ceglio, *Lawrence Livermore National Laboratory, Livermore, CA.*

Recent advances in the field of soft X-ray optics, including interference mirrors, splitters, lenses, gratings and picosecond detectors, will be reviewed.

11:30 AM TuB3 (Invited Paper)

Population Density and VUV Gain Measurements in Laser-Produced Plasmas, R. H. Dixon, J. F. Seely, and R. C. Elton, *Naval Research Laboratory, Washington, DC.*

Population densities and vuv gain coefficients are deduced from absolute resonance line intensities in laser-produced plasma ions and compared with analytical pumping models.

12:00 M TuB4 (Invited Paper)

Pulsed Hollow Cathode Discharge for XUV Lasers and Radiation Sources, R. W. Falcone, D. E. Holmgren, and K. D. Pedrotti, *Edward L. Ginzton Laboratory, Stanford University, Stanford, CA.*

High densities of excited state lithium ions have been created in a novel pulsed hollow cathode discharge. The discharge may be useful for the realization of proposed schemes for XUV lasers and radiation sources.

12:30 LUNCH

TUESDAY, MARCH 9 Continued

MULTIPHOTON PROCESSES I

S. Smith, *President*

2:00 PM TuC1 (Invited Paper)

Multiphoton Ionization of Atoms and Molecules, R. N. Compton and John C. Miller, *Oak Ridge National Laboratory, Oak Ridge, TN.*

We will discuss recent experimental results on multiphoton ionization of atoms (rare gas and alkali) and small molecules (NO , I_2 , H_2S , and NH_3) at low ($\sim 10^{-6}$ torr) and high (~ 100 torr) pressure.

2:30 PM TuC2 (Invited Paper)

First Observation of Photoionization of Laser-Excited Atoms by Synchrotron Radiation, J. M. Bizau, F. Willeumier, P. Dhez, D. Ederer*, J. L. Legouet**, J. L. Picque**, and P. Kocht, *Lab. Spectroscopy Atomique et Ion, and LURE, Orsay, France.*

Photoionization of laser-excited atoms in the VUV has been studied using monochromatized synchrotron radiation to ionize the atoms and an electron spectrometer to analyze the photoelectrons.

*National Bureau of Standards, Washington, DC.

**Lab. Aime, Cotton, Orsay, France.

†Yale University, New Haven, CT.

3:00 PM TuC3

Electron Spectra of Four-Photon Ionization of NO, P. Kruit, J. Kimman and M. J. Van der Wiel, *FOM-Institute for Atomic and Molecular Physics, Amsterdam, The Netherlands.*

We report extreme non-Franck-Condon vibrational populations of NO^+ formed by four-photon absorption to autoionizing states via an intermediate two-photon resonance.

3:15 PM TuC4

Generation of Narrow-Band Tunable Radiation in the 1200 Å Spectral Region, F. S. Tomkins and R. Mahon*, *Argonne National Laboratory, Argonne, IL.*

Work on frequency up-conversion in Hg vapor has been extended further into the VUV, producing continuously tunable, narrow-bandwidth radiation from 1220 Å to 1197 Å.

*Department of Physics and Astronomy, University of Maryland, College Park, MD.

3:30 PM REFRESHMENT BREAK

TUESDAY, MARCH 9 Continued

MULTIPHOTON PROCESSES II

W. H. Parkinson, *President*

4:00 PM TuD1 (Invited Paper)

Multistep Excitation of Alkaline Earth Atoms to Autoionizing States, T. F. Gallagher, *Molecular Physics Laboratory, SRI International, Menlo Park, CA.*

A multistep laser excitation technique is described which has been used to excite atoms to states as high as 10 eV above the atomic ground state.

4:30 PM TuD2 (Invited Paper)

Vacuum Ultraviolet Spectroscopy of Molecules Using Third-Harmonic Generation in Rare Gases, John C. Miller, C. D. Cooper, and R. N. Compton, *Oak Ridge National Laboratory, Oak Ridge, TN.*

Absorption and one- and two-photon ionization studies in the vacuum ultraviolet have been performed using tunable, narrow bandwidth light produced by third-harmonic generation in rare gases.

NEW YORK—AMERICAN EXCHANGE

8:00 PM POST DEADLINE POSTER PAPER SESSION

WEDNESDAY, MARCH 10

MIDWEST—PACIFIC EXCHANGE

NON-LINEAR MIXING I

W. C. Martin, *President*

8:30 AM WA1 (Invited Paper)

Non-Linear Mixing, Karl H. Welge, *Fakultät für Physik, Universität Bielefeld, West Germany.*

We report multiphoton excitation — ionization investigations with tunable VUV laser radiation applied to atomic hydrogen. Objectives are: (1) high Rydberg state excitation, (2) polarized proton production and (3) Doppler spectroscopy of H and D atoms.

9:00 AM WA2 (Invited Paper)

Generation of Coherent Radiation Below 100 nm in Hg Vapor, R. F. Freeman and J. Bokor*, *Bell Laboratories Murray Hill, NJ.*

Tunable coherent light below 100 nm has been produced via 4-wave mixing in Hg vapor. Dynamics of two-photon saturation on the power output was investigated.

*Bell Laboratories, Holmdel, NJ.

9:30 AM WA3 (Invited Paper)

Spectroscopy of CO and NO Using Coherently Generated VUV, C. R. Vidal, *Max-Planck-Institut für Extraterrestrische Physik, Garching, West Germany.*

10:00 AM COFFEE BREAK

WEDNESDAY, MARCH 10 *Continued*

NON-LINEAR MIXING II

H. Pilloff, *President*

10:30 AM WB1 (Invited Paper)

Generation of Narrowband Tunable VUV Radiation, R. Hilbig and R. Wallenstein, *Fakultät für Physik, Universität Bielefeld, Bielefeld, West Germany.*

Narrowband VUV radiation is generated by resonant and nonresonant frequency conversion of dye laser light in mercury and in the rare gases Xe and Kr.

11:00 AM WB2 (Invited Paper)

Generation of Tunable, Coherent 79 nm Radiation by Frequency Mixing, H. Egger, T. Srinivasan, K. Boyer, H. Pummer and C. K. Rhodes, *Department of Physics, University of Illinois at Chicago Circle, Chicago, IL.*

Tunable coherent radiation in the 79 nm range (200 mW) has been generated by sum frequency mixing of two ArF* photons and one visible dye laser photon.

11:30 AM WB3 (Invited Paper)

Upconversion of Laser Radiation to γ -Ray Energies, C. B. Collins, *Center for Quantum Electronics, University of Texas at Dallas, Richardson, TX.*

We review the general problem of multiphoton processes which might occur at the nuclear level when one of the photons belongs to an intense radiation field from a laser at optical frequencies.

12:00 M MEETING ADJOURNED.

HIGH RESOLUTION SPECTROSCOPY I

High Resolution Spectroscopy

Thomas J. McIlrath
Institute for Physical Science and Technology
University of Maryland
College Park, MD 20742

High resolution spectroscopy in the vacuum ultraviolet region has, until recently, been limited to work with long focal length grating spectrographs. The best resolution with state-of-the-art instrumentation is approximately 250,000, but the photographic method provides simultaneous measurement of $\approx 3 \times 10^4$ data points with modest exposure times. The development of tunable coherent sources in the VUV promises to increase the available resolution by well over an order of magnitude, but tunable coherent sources involve sequential point-by-point data collection with typical scanning ranges of $\approx 10^2 \text{ cm}^{-1}$.

The inherent atomic or molecular linewidth in the VUV (in cm^{-1}) is often greater than in the visible region. The natural linewidth of a level connected to the ground state with unit oscillator strength is $\approx 0.04 \text{ cm}^{-1}$ for a 1000\AA resonance line compared with $\approx 0.001 \text{ cm}^{-1}$ for a 6000\AA line. Higher atomic levels and many molecular levels have narrower inherent widths while autoionizing or predissociating levels are broader and are often resolved by conventional means. Unless low temperature beams or non-linear narrowing techniques are used, Doppler broadening often determines the maximum useable resolution. Doppler widths are $\approx 1 \text{ cm}^{-1}$ for H (1215\AA) at room temperature and $\approx 0.09 \text{ cm}^{-1}$ for Xe (1295\AA). In general, lasers with high resolution and limited spectral coverage complement grating instruments with moderate resolution and extended coverage.

SUMMARY

TUNABLE, COHERENT SOURCES FOR
HIGH-RESOLUTION VUV AND XUV SPECTROSCOPY

B. P. Stoicheff

Department of Physics, University of Toronto
Toronto, Ontario, M5S 1A7, Canada

About 20 years ago, great strides were made in high-resolution spectroscopy in the VUV with the building of large grating spectrographs which provided resolving powers of $\sim 3 \times 10^5$. To-day, with the use of tunable laser sources, resolving powers of $\sim 10^7$ have been reached in limited wavelength ranges, with $\sim 10^5$ available over most of the VUV and XUV regions to as short as ~ 50 nm.

A review of tunable laser sources for VUV and XUV spectroscopy will be presented, with emphasis on the resolution achieved to date. These results will be illustrated with recently obtained spectra.

SPECTROSCOPIC MEASUREMENTS OF SUB-LASER
LINEWIDTH AUTOIONIZATION RATES

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If the lifetime of an autoionizing state is much shorter than the pulse length of the laser exciting it, then it is possible, with sufficient laser power, to ionize the entire atomic population. When the laser is tuned off resonance, it may still have sufficient power to ionize almost the entire population. The detuning at which the ionization efficiency will drop to 50% depends on the product of the laser pulse's energy density and the excited state's autoionizing rate.

We have used this relationship to measure small autoionization rates of some 6Pns states of barium. By increasing our laser power until the depletion broadened lineshape was larger than our laser's linewidth, we were able to determine autoionization-induced linewidths that were even smaller than our laser linewidth.

The laser energy density required for these measurements was quite modest, $\sim 100 \mu/\text{cm}^2$ could be used to induce a depletion-broadened linewidth of several cm^{-1} . This suggests that this effect is not only a useful measurement technique but also may produce unexpected broadenings, resulting in incorrect linewidth measurements.

LASER-INDUCED IONIZATION OF Na VAPOR:

A NEW MECHANISM FOR PRODUCING Na_2^+

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The production of Na_2^+ ions by off-resonant laser excitation in the 5800-6200 Å region mainly results from two-photon absorption by the Na_2 molecule to highly excited *gerade* states followed by (a) direct ionization by absorbing a third photon or (b) coupling to the molecular Na_2 $D^1\Pi_u$ Rydberg state which is subsequently ionized by absorbing a third photon. This mechanism, i.e., a two-photon resonance three photon ionization process, explains a recent experimental observation of Roussel et al.^[1] It is suggested that the very same mechanism is also responsible for a similar observation reported by Polak-Dingels et al.^[2] in their work using two crossed Na beams. In the latter two studies the laser-induced associative ionization processes were reported to be responsible for producing the Na_2^+ ion. From the ratio of molecular to atomic concentration in the crossed beam experiment [2] we estimate that the cross section for producing Na_2^+ through laser-induced associative ionization is at least four orders of magnitude smaller than ionization through the two-photon resonance three photon ionization process in Na_2 molecules.

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HIGH RESOLUTION SPECTROSCOPY II

Optical-Optical Double Resonance Multiphoton Ionization Spectroscopy
of NO.

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Resonantly enhanced multiphoton ionization of NO has been investigated using two tunable dye laser beams oppositely directed and having a common focus. The focus lies between two electrodes in a cell containing 4-15 torr of NO. The first laser beam pumps a rovibronic level of either the A or C state by a two or three photon process. The second laser is scanned over a spectral region leading finally to ionization by one or two photons sometimes assisted by field or collisional ionization. We have obtained high resolution data on high Rydberg and non-Rydberg states below and above the ionization limit with special attention to the Rydberg-non-Rydberg perturbations and dynamic processes such as autoionization, predissociation, collisional and field ionization and rotational relaxation. The spectral simplification resulting from the rotational selectivity of the double resonance technique has yielded easily analyzed spectra in cases for which the ordinary one-photon spectrum is hopelessly congested or overlaid by strong, broadened lines. One of many examples is the strongly perturbed $v=8$ level of the L valence state. Decay processes can yield either peaks or dips often with measurable widths. Thus the $N=1, v=1$ level of the $5p\sigma$ state, which is obscured in the one-photon spectrum is found to have a predissociation width of about 6cm^{-1} . In contrast, the $v=3$ levels of the 4f, 4d, 6s and 7s Rydberg states, all of which lie above the ionization limit for $v=0$ or $v=1$ and hence are subject to vibrational

autoionization, are found to have widths $< .5\text{cm}^{-1}$ in agreement with our calculations. We have studied linear and quadratic, static and dynamic Stark effects as well as pressure shifts and broadening of Rydberg states with values of n up to ~ 35 . Pulse delay studies have shown adiabatic following of dressed states under conditions of strong power broadening and A.C. Stark shifts. Resonant structures which appear as either peaks or dips as a function of laser power are explained in terms of Fano theory.

FREQUENCY CONVERSION PROCESSES IN HYDROGEN

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Multiphoton Raman-like transitions taking place above and below the ionization threshold of an atom can be useful for generating energetic photons of well-defined energies. In these processes, the atomic electron makes a bound-bound transition. It first absorbs N photons of the incoming light and then reemits a photon whose energy depends on N and on the discrete final state.

A quantitative analysis of such processes in Hydrogen is presented. The most favourable conditions for observing the emitted light are determined by using accurate calculation techniques. By comparing transitions of same perturbation orders, we show that non-resonant frequency conversion is more efficient when the energy of N photons exceeds to a large extent that of ionization.

The case of resonances resulting from the interaction between the atom and the laser field is discussed.

We show how particular resonances, occurring when the energy difference between two excited atomic levels is equal to the photon energy, can appear only in frequency conversion processes. Such resonances allow considerable enhancements of frequency conversion efficiency while keeping the ionization rate unchanged. On the other hand, the ground state can be resonantly coupled to an excited state by several quanta. Since these two kinds of resonances can occur simultaneously, it is possible to find experimental situations proper to generate ultra violet signals of reasonable intensities from tunable lasers of moderate powers.

Atomic H and D Concentrations and Velocities Measured with
Harmonically Generated Lyman- α (1215 Å) Radiation

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A resonance fluorescence scattering system remotely senses H- and D- atom concentrations in the 10^9 cm^{-3} range in a tokamak. The Doppler broadening measures temperatures down to 100°K.

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LASER INDUCED ENERGY TRANSFER IN HIGHLY EXCITED STATES IN MOLECULES

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Processes involving high-energy molecular states are experimentally demonstrated using multiphoton excitation techniques.

The first observation of laser-assisted intermolecular energy transfer between electronic states of carbon monoxide is reported. Cross sections for such processes involving high-energy ($E \sim 11$ eV) Rydberg states are shown to be larger than 10^{-16} cm² at the laser intensities approaching 10^{10} W/cm². As predicted by theory, at such high intensities, the originally linear in P/A target state fluorescence intensity dependence goes over to $(P/A)^{1/2}$ dependence. Our experiments clearly illustrate the care necessary in interpreting multiphoton spectra taken at high laser powers.

In molecular nitrogen selective excitation of the $b'^1\Sigma_u^+$ valence state ($E \sim 13$ eV) was achieved. The excitation process utilizes a two-photon transition to the intermediate $a'^1\Pi_g$ state followed by a single-photon transition to the target state. The excitation is confirmed by observation of the VUV emissions from the b' state to high-vibrational levels of the ground molecular state. The b' state population of N₂ is estimated to be greater than 10^{13} cm⁻³. Generation of highly excited atomic nitrogen following the two-photon excitation of the $a'^1\Pi_g$ molecular state is reported and is interpreted as a collisional process involving metastable atoms and molecules.

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MB5-1

An Overview of Lithium Spectroscopy

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DIRECT GENERATION I

DIRECT GENERATION OF XUV RADIATION*

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The technology of high spectral brightness rare gas halogen (RGH) excimer lasers can be readily used to generate bright coherent radiation in the extreme ultraviolet (XUV) range. As demonstrations of this capability, tunable (XUV) radiation at ~ 64 nm and ~ 79 nm has been generated using a high spectral brightness ArF* (193 nm) source by frequency tripling and frequency mixing, respectively. In the case of tripling, it has been experimentally determined that the conversion efficiency is limited by photoabsorption of generated XUV photons by the nonlinear medium. It is shown that a simple flow geometry that confines the nonlinear medium to the conversion zone by a buffer gas of low absorption cross-section sharply reduces these losses and leads to correspondingly enhanced conversion efficiencies. With this apparatus, at 64 nm, 30 W, 10 nsec pulses have been generated corresponding to a conversion efficiency of 10^{-6} . This upconverted radiation is tunable over 0.2 nm, the range determined by the gain profile of ArF* amplifiers. Broad tunability is achieved with the use of a dye laser in a process of frequency mixing. Tunable radiation with peak powers of 200 mW at ~ 79 nm has been generated by frequency mixing 2 ArF* photons and one visible dye photon (420 - 460 nm). Since nonlinear processes generally favor the use of short pulses, 10 psec pulses at 193 nm have been generated by frequency tripling of a pulse amplified, synchronously pumped mode-locked dye laser. Amplified radiation of this source is used to obtain coherent radiation at 64 nm with a power of 20 kW by frequency tripling, a value corresponding to an enhanced conversion efficiency of $\sim 10^{-5}$. An analysis of the expected limiting performance of RGH systems for the production of XUV and soft x-rays will be given.

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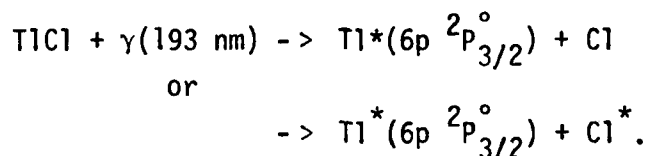
ANTI-STOKES RAMAN LASERS

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This paper reports what we believe to be the first observations of stimulated, anti-Stokes Raman laser emission. A metastable $\text{Tl}^*(6p\ ^2P_{3/2}^\circ)$ population inversion was created by selective photodissociation of TlCl , and Raman scattering using 532 nm and 355 nm pump lasers resulted in stimulated emission at 376 nm and 278 nm, respectively. Likewise, inverted $\text{I}^*(5p\ ^5P_{1/2}^\circ)$ atoms have been used as a Raman medium; and with a 206 nm pumping source, we have generated tunable VUV radiation at 178 nm.

Absorption by a TlCl molecule of an ArF^* excimer laser photon at 193 nm results in photodissociation along the paths



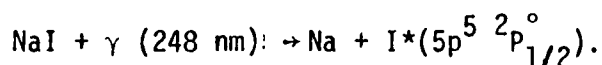
Since no $\text{Tl}(6p\ ^2P_{1/2}^\circ)$ ground state atoms are produced by this process, the $6p\ ^2P_{3/2}^\circ$ metastable state is inverted with respect to ground. Subsequent absorption of a strong pumping field tuned near a dipole allowed state with opposite parity may connect the metastable and ground states via a two-photon, anti-Stokes Raman scattering process.

Stimulated anti-Stokes emission was observed at a 532 nm pump energy of about 15 mJ. Accounting for the stimulated emission from both ends of the cell, a pulse energy at 376 nm of 1.8 mJ was measured. The conversion efficiency may be defined as the ratio of the anti-Stokes output energy to the 193 nm

energy absorbed in the active volume. Approximately 26 mJ of the 193 nm pump was absorbed in the active volume, implying an efficiency of ~7 percent. The observed output energy implies a $\text{Tl}(6p\ ^2P_{3/2}^{\circ})$ storage density of approximately 4×10^{16} atoms/cm³ was created by the photodissociation step.

Anti-Stokes Raman emission at 278 nm using the 355 nm pump wavelength was also observed. With an input energy of ~100 mJ/pulse at 355 nm, the output energy at 278 nm from both cell ends was approximately 2.5 mJ/pulse. The conversion efficiency in this case was 10 percent.

In a second experiment, an $\text{I}^*(5p\ ^5P_{1/2}^{\circ})$ inversion was created by selective photodissociation of NaI using a KrF^* excimer laser, via the reaction



Pump radiation to drive the Raman process was generated at 206 nm by Raman scattering a frequency doubled dye laser in a high pressure H_2 cell. The third anti-Stokes of this process at 206 nm was collected and overlapped spatially with the 248 nm laser in the NaI oven.

At a 206 nm input energy of 50 μJ , approximately 35 μJ (i.e., 7 kW) of 178 nm radiation was generated and was tunable over $10\ \text{cm}^{-1}$. The 206 nm pump laser could be delayed as much as 500 nsec after the arrival of the KrF^* dissociation laser and still maintain stimulated emission at 178 nm. The observed output energy implies that an I^* inversion density of 6.2×10^{14} atoms/cm³ was created in the dissociation step, and a conversion efficiency of 1.7 percent was measured.

Anti-Stokes Raman lasers, due to their tunability and relatively high conversion efficiency could be useful for upconverting a variety of laser sources. Upconverters for CO_2 lasers could be constructed in alkali atoms to

yield radiation in the 400 nm range. In addition, the use of other halogen or metal atoms may permit the construction of tunable VUV Raman lasers in the 100 nm to 200 nm range.

RECOMBINATION LASERS IN THE VACUUM ULTRAVIOLET

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Isoelectronic scaling of known laser transitions to higher ion stages¹ has recently been shown to be a useful technique for scaling electron-ion recombination lasers, produced in expanding plasmas, to shorter wavelengths.² The observation of laser action on a sequence based upon the 4f-5d transition in Ag I (1832 nm), Cd II (537.8 nm) and In III (298.3 nm) is perhaps the clearest example of this technique to date.² Another sequence, based upon the 4f-3d transition in Li I (1870 nm) (recently observed in laser action in a segmented plasma arc³ device) and Be II (457.4 nm) (reported earlier⁴) suggests possible new recombination lasers in B III (207.7 nm) and C IV (116.8 nm).

Increased gain is expected on isoelectronic transitions of higher ionization stages. This expectation is based primarily upon the increased electron density that can be allowed without thermalizing the relevant laser levels. Such gain scaling conclusions suggest potential gains ranging from $.03 \text{ cm}^{-1}$ to 15 cm^{-1} at 116.8 nm in C IV. The smaller number, based upon scaling of experimentally observed gain in neutral atom and single ion recombination lasers, should be considered a lower limit; whereas the larger number, based upon a H-like plasma model that has not been confirmed experimentally, is probably overly optimistic.

Using the more pessimistic estimate of gain, 10 carbon plasmas (of the appropriate temperature and density) in a row should produce a minimum of 30% gain and allow laser oscillation at 116.8 nm using state-of-the-art laser mirrors at that wavelength. Such a laser should be capable of producing approximately 50 mJ/pulse in the vacuum ultraviolet.

To test these ideas experimentally, 10 laser-produced carbon plasmas will be excited transversely with respect to the optical cavity by placing carbon targets at the focus of 10 identical 2 J CO_2 laser beams (each having a pulse duration of $\sim 70 \text{ ns}$). The carbon plasmas will be allowed to expand into the region of the optical cavity in

the presence of a low-pressure background gas of He to produce cooling and thereby initiate rapid recombination. Two MgF_2 overcoated 70% reflecting Al mirrors will provide optical feedback with one mirror allowing partial transmittance through its LiF substrate.

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DIRECT GENERATION II

ANTI-STOKES SCATTERING AS AN XUV RADIATION SOURCE

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SUMMARY

The paper will describe the use of anti-Stokes scattering as a radiation source for high resolution spectroscopy in the 100 Å to 1000 Å spectral region, and as a flashlamp for the construction of XUV lasers.

The radiation source is based on spontaneous anti-Stokes scattering of incident laser photons from excited metastable atoms in an electrical discharge. The frequency of the scattered photons is equal to the sum of the metastable storage frequency and the frequency of the incident laser photon. The radiation has the same time duration as the incident laser and may thus readily be of picosecond time scale. It is polarized, has a linewidth equal to that of the convolution of the laser linewidth and Doppler width of the emitting species, and can be tuned by tuning the incident laser frequency. In recent experiments we have demonstrated that the intensity of the tunable anti-Stokes radiation is sufficiently great that it may be distinguished from the background radiation of the plasma, and that therefore no monochromator need be used to obtain absorption spectra. We report high resolution spectra of potassium in the region of 550 Å.

We will describe experiments aimed at using the anti-Stokes scattering to make a picosecond time scale flashlamp at 199 \AA ; and then using this flashlamp to construct a 200 \AA laser. In recent experiments a pulsed hollow cathode discharge has been used to obtain storage populations in the metastable lithium ion of $5 \times 10^{11} \text{ ions/cm}^3$. These populations correspond to a 199 \AA flashlamp power density of about 10^4 W/cm^2 . The target state for the proposed laser is a doubly excited state of neutral lithium which is prohibited by selection rules from autoionizing. Calculations indicate that we will be able to obtain a gain of about 10% per cm on the 207 \AA transition of neutral lithium.

PHOTO-AUTOIONIZATION PUMPED BA ION LASER

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We have studied the selective autoionization process $6p_{3/2}np \rightarrow 6p_{1/2} + e^-$ in atomic Ba, and have achieved inversion in Ba^+ via this process. Using two lasers, neutral Ba is first excited from its $6s^2 \ ^1S_0$ ground state to a Rydberg state $6snp \ ^1P_0$, with a tunable laser near 247 nm. Another dye laser, near 455 nm, further excites the Ba to $6p_{3/2}np$, whereupon the atom autoionizes. We have studied the excitation spectrum as a function of the wavelength of the second laser, and have found it similar to that observed in Ba $6p_{3/2}ns$ states.¹

The dominant decay channel is found to be autoionization to the $6p_{1/2}$ excited state of Ba^+ . Dramatic evidence for this behavior is provided by the observation of laser action in Ba^+ on the $6p_{1/2} \rightarrow 6s_{1/2}$ transition at 493 nm and the $6p_{1/2} \rightarrow 5d_{3/2}$ transition at 650 nm. This decay channel involves a quadrupole autoionization, and

between the fine structure states of the ion it overcomes the dipole autoionization to the ground state. This process is related to the two-step autoionization process observed in a number of atomic systems at much higher energies using electron impact excitation.²

In this work, it was found that the visible laser used in the two-step preparation of the $6p_{3/2}np$ state could be tuned far enough off exact resonance so that it could simultaneously saturate the $6snp - 6pnp$ transition, and drive the $6p_{1/2} - 6f_{5/2}$ transition in the ion. Under these conditions, an intense 4-wave mixing output was observed on the $6f_{5/2} -- 6d_{3/2}$ transition in Ba^+ , where two input waves are derived from the above visible laser, and one wave arises from the internal laser generated in the Ba^+ along the $6p_{1/2} -- 5d_{3/2}$ transition by the selective autoionization mechanism. The output wavelength is at 167.4 nm, and we estimate the output power to approach 1KW/pulse.

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Stimulated Vacuumultraviolet Emission From Rare Gas Crystals

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The advantages of solid state rare gas lasers compared to the gas phase lasers can be a high excimer density at reduced losses from excimer quenching and an efficient energy deposition. Free standing optically clear and 2 cm long rare gas crystals have been excited by an intense electron pulse (400 keV, 3kA, 3×10^{-9} s, 3 J). Densities of excited centers of 10^{17} to 10^{18} cm^{-3} have been created in the crystals without reduction of the optical quality. The crystals emit the well known excimer bands at 128 nm (Ar), 145 nm (Kr) and 160 - 180 nm (Xe). Each band consists of two components, the dipole allowed transition $^1\Sigma_u^+ \rightarrow ^1\Sigma_g^+$ which is responsible for amplified stimulated emission and a long living transition $^3\Sigma_u^+ \rightarrow ^1\Sigma_g^+$.

The Ar_2^* band at 128 nm is emitted with more than 10^{16} photons per pulse. Amplified stimulated emission is indicated by a reduction of the fwhm of the band, by radiative decay of most of the $^1\Sigma_u^+$ population within the excitation pulse width of 3×10^{-9} s and by an progressive increase of the intensity emitted into 10^{-2} sterad with excitation energy. In Xe and Kr crystals it will be more difficult to achieve stimulated emission because the emission yield at 11 K is low due to strong selfquenching of the excimers. The yield for Xe and Kr increases at higher temperatures but on the expense of crystal quality. Xe and Kr crystals show a prompt continuum emission starting below 400 nm and extending beyond 1000 nm which may originate from a hot electron plasma.

POSTER SESSION

LASER-INDUCED CONTINUUM STRUCTURE IN
MULTIPHOTON IONIZATION SPECTROSCOPY

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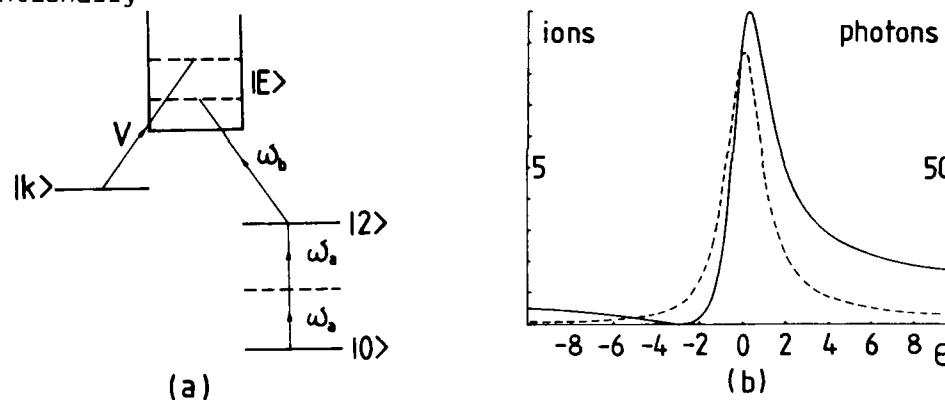
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Multiphoton processes with significant continuum contributions are enhanced by exploiting autoionizing resonances embedded in the continuum by configuration interaction (1). Tunable "embedding" can be induced by radiative coupling from an initially empty discrete state; such "pseudoautoionizing" states can be shifted and broadened at will.

Transitions from state $|a\rangle$ to the dressed continuum $|F\rangle$ are described by $V_{aF} = \langle a | V | F \rangle = V_{aE} (q_a + \epsilon) / (\epsilon - i)$ where q_a is the Fano parameter and ϵ the detuning of E from the embedded state in units of the width of $|a\rangle$. Here q_a is the ratio of the real part of the two-photon Rabi frequency from $|a\rangle$ to the embedded state to the imaginary part of this frequency; recent measurements report $q \sim 7$ for caesium transitions (3).

We have investigated the use of such embedding in several multiphoton processes. Fig (a) is a level-scheme for two-photon resonant three-photon ionization to a dressed continuum and (b) the enhanced ionization and up-conversion rate $|x^{(3)}|^2(\text{photon})$ versus detuning from the induced structure for $q_0 = 2.5$, $q_2 = 3$. The ionization rate can be increased by one order of magnitude and the up-conversion rate by two orders of magnitude. We will report power broadening and Rabi modulation in structured continua. Such structures are often present unintentionally



in multiphoton experiments but could be profitably exploited,

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Two-photon Excitation of Argon

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Multiphoton excitation is describable in terms of rate equations^{1,2} when the generalized n-photon Rabi frequency and the excited-state photo-ionization rate are much less than the laser bandwidth. This is generally the case when powerful excimer lasers are used to excite atomic levels in rare gases. The two photon excitation rate may be written as

$$W = \alpha I_1 I_2 G(\omega_1, \omega_2) \quad , \quad (1)$$

where ω_1 and ω_2 are the frequencies of lasers 1 and 2 respectively, I_1 and I_2 are the laser intensities, $G(\omega_1, \omega_2)$ is the bandwidth-dependent line-shape function and α is the two-photon excitation parameter defined by Eq. (1).

We calculate the two-photon excitation parameter for the ground state to $3p^5 4p$ 1S excited state transition in argon. In the non-relativistic dipole-length approximation α may be written (in atomic units) as

$$\alpha = \frac{8\pi^3}{c^2} \left| \sum_n \langle \psi_f | \sum_{i=1}^N \epsilon \cdot \vec{r}_i | \psi_n \rangle \langle \psi_n | \sum_{i=1}^N \epsilon \cdot \vec{r}_i | \psi_i \rangle \right| \times [(E_i - E_n + \omega_1)^{-1} + (E_i - E_n + \omega_2)^{-1}]^2 \quad , \quad (2)$$

where c is the speed of light, $\hat{\epsilon}$ is the direction of radiation-field polarization and \sum_n represents a sum over bound states and an integration over the continuum. We use multiconfiguration Hartree-Fock wave functions to evaluate the matrix elements found in Eq.(2). Comparison is made with average-energy closure methods based on other atomic models.

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Direct Observation of Hot Electron Spectra fromLaser Illuminated Sodium Vapors. J.L. LEGOUET, J.L.

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Low intensity CW laser was tuned to D_2 line in sodium. Large numbers of electrons with kinetic energy 4.2 and 5.4 eV were observed.

Low intensity (few W/cm^2) cw ring dye laser photons tuned to the D_2 line (589 nm) crossed a weakly collimated Na atomic beam with density ~ 5 -20% to the $Na(3^2P_{3/2})$ level. A cylindrical mirror analyzer was used to record spectra of hot electrons emitted from the interaction volume with kinetic energy $E > 3$ eV; the two most prominent peaks were observed near 4.2 and 5.4 eV. Their energies and relative intensities support strongly a multi-step collisional ionization process in which associative ionization, energy pooling, and collisional ionization of higher excited states produced primary electrons. Then the super-elastic collisions with $Na(3p)$ atoms raised their energy to the observed peak values.

*Work carried out at LURE, Univ. Paris Sud, Orsay, France.

2π Electron Spectrometer for Multi-Photon Ionization StudiesP. Kruit⁺, F.H. Read^{*} and M.J. Van der Wiel⁺⁺FOM-Institute for Atomic and Molecular Physics,

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In highly focussed nanosecond laser pulses the yield of electrons must be kept below about one hundred per lasershot, otherwise the space charge of the ions in the laser-focus will seriously influence the electron energy spectrum. Especially for the study of electrons produced in multiphoton ionization a spectrometer has been designed which accepts an opening angle of 2π sr. The laser beam is focussed in a region where a magnetic field of 1 T is present. The strong field diverges along the axis of a 50 cm time-of-flight tube to a weak field of 10^{-3} T, in which an electron detector is positioned. Photoelectrons with any velocity in the direction of the detector spiral around the magnetic field lines and reach the detector. In the region of the field divergence the spread in forward velocities is reduced to 5×10^{-4} by an inversed magnetic-mirror principle. The total velocity is unaffected. Experiments show the resolution to be 20 meV for electrons up to 10 eV energy. By applying a retarding potential in the high magnetic field region it is possible to select an opening angle different from 2π sr, thus permitting measurements of angular distributions as well.

Autoionizing and High-Lying Rydberg States of Lutetium Atoms

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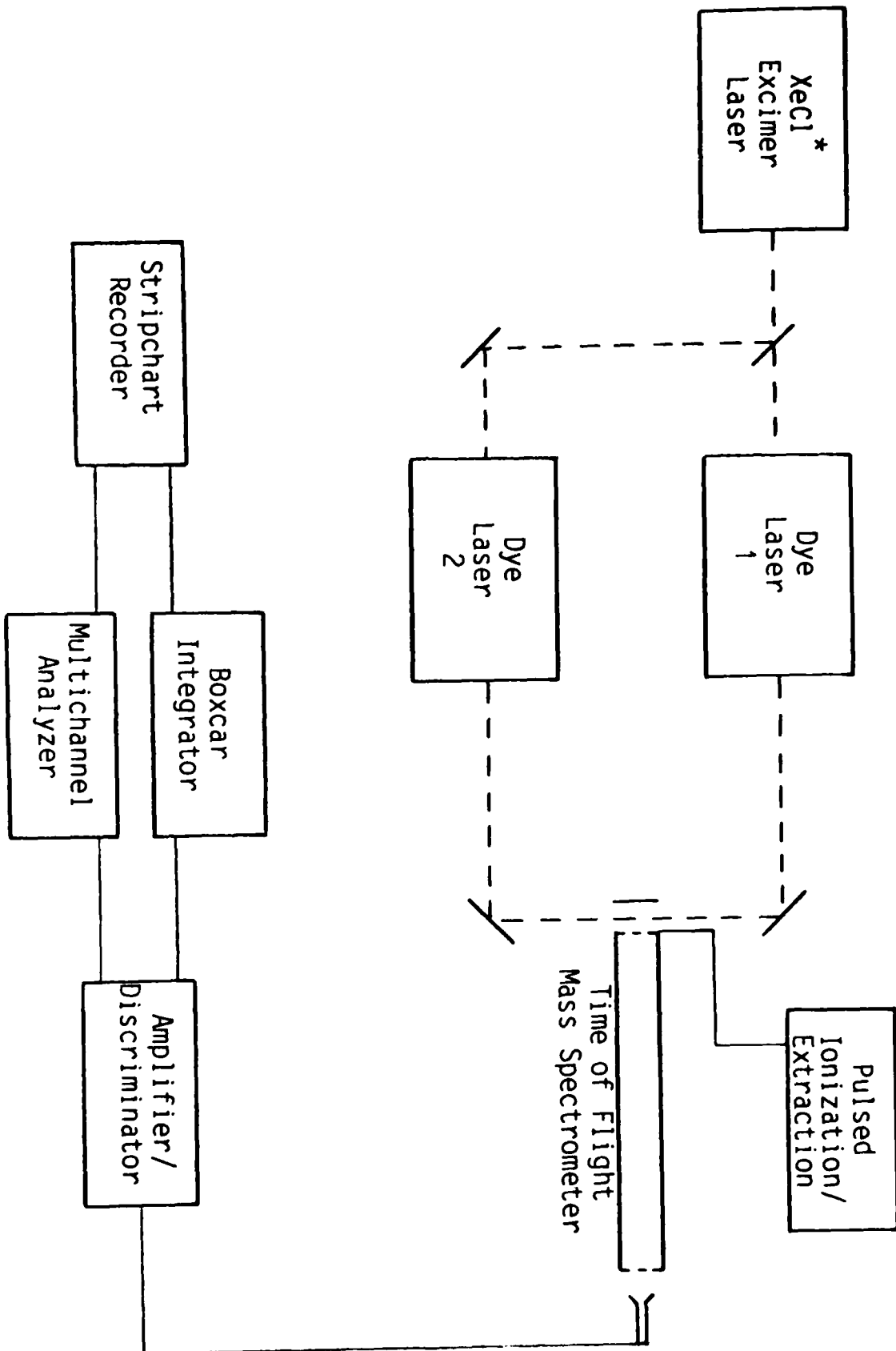
SUMMARY

A pulsed-field time-of-flight mass spectrometer was used in conjunction with two XeCl* excimer laser pumped dye lasers to examine the spectroscopy of Lu atoms near the 5.4 eV ionization limit, Fig. 1. A fixed frequency laser, 22125 cm^{-1} , was used to saturate the $5d6s^2\ ^2D_{3/2} \rightarrow 5d6s6p\ ^2D_{3/2}$ transition, while a frequency scanned laser was used to probe transitions from the excited state to high Rydberg or low lying autoionizing levels. Rydberg levels close to the continuum were observed and assigned, as were several autoionizing features not previously reported.^{1,2}

In addition, a parametric study of the effects of laser intensity and optical delay were carried out and extensively modeled in terms of a rate equations formalism for the multiphoton ionization process. Implications of these results for the detection of ultratrace quantities of Lu will be discussed.

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High Average Brightness Rare-Gas
Halide Laser Technology

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Summary

Discharge pumped rare-gas halide lasers are attractive pump sources for non-linear UV generation techniques such as multiwave mixing and Raman conversion because of their high peak power. To extend the performance of these lasers to much higher average powers, we have used high velocity closed loop gas flow systems that allow both 100 and 1000 Hz operation at average powers ranging from 1 to greater than 100 Watts at 308 nm. At the highest powers, active heat exchangers and sidewall acoustic wave dampers using lossy materials are required. Careful design of flow bends and the laser head interface is also required to prevent flow separation.

For nonlinear conversion brightness is more relevant than power for describing the usefulness of a given laser. Using a positive branch unstable resonator ($M=4$), we measure far field beam quality between 3 and 4 times diffraction limited at greater than 30 Watts, or average brightness of 5×10^8 W/(cm²-steradian). In a smaller device, beam divergence of about 1.5X diffraction limited at 0.25 Watt has been shown, producing average brightness of 2×10^7 W/(cm²-steradian). Preliminary data shows that this improved divergence can be transferred to the higher power device using injection locking techniques, offering the possibility of perhaps 10X improved brightness.

Analysis of vuv and Soft x-ray Lasing

Based on Charge-Exchange Mechanisms

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We report theoretical investigations of long-wavelength laser mechanisms based on efficient formation of electronically excited atoms or ions by charge exchange. In one promising practical arrangement a plasma containing the ions clashes with a vapor containing the neutral-atom collision partners. If charge transfer wins over competing photo-chemical processes, then inversion of population in an electronically excited state appropriate for lasing results. We present a systematic procedure for determining whether and under what conditions lasing occurs for given collision partners. Via rate equations describing ionic and neutral populations we assess in a first approximation the roles of charge transfer, spontaneous emission from the excited specie, and plasma and vapor pulse shapes in achieving population inversion. We next investigate basic laser action by extending the rate equations to include stimulated emission and photoionization of the excited specie by laser radiation. Our refined analysis is based on detailed studies of the other photo-chemical processes to determine their relative effects on laser action. Where necessary, we perform cross-section calculations for various processes to assess their importance.

RESONANTLY ENHANCED MULTIPHOTON IONIZATION AND THIRD-HARMONIC GENERATION IN CARBON MONOXIDE GAS*

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Multiphoton ionization (MPI) and third-harmonic generation (THG) in gaseous carbon monoxide (CO) have been studied using photons generated from a Nd:YAG-pumped tunable dye laser (Quanta-Ray). The MPI and THG were studied in a static-gas ionization cell with a vacuum ultraviolet (VUV) radiation detector as shown in Fig. 1. In both experiments the dye laser output was focused to give a power density on the order of 10^9 W/cm^2 .

Resonantly enhanced MPI signals through the $A^1\Pi \leftarrow X^1\Sigma$ (fourth positive system) transition of CO disappear at wavelengths to the blue of the R-head in the $v' = 0, 1, 2$, and 3 levels; concurrently, intense VUV third-harmonic radiation is detected in the forward direction with respect to the pump laser. The MPI and THG signals are shown in Fig. 2 for the $v' = 1$ and $v' = 2$ at low dispersion. Shown in Fig. 3 is absorption of the VUV by the $\tilde{B} - 9$ level of

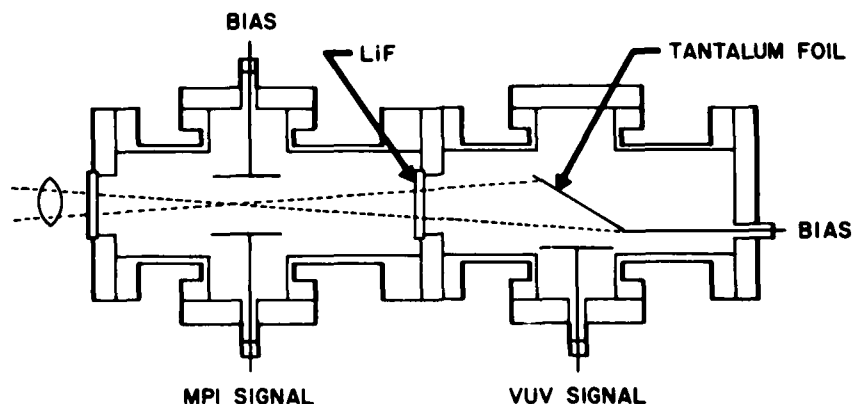


Fig. 1. Schematic diagram of the apparatus used to measure the MPI and the third-harmonic VUV radiation. The pump laser can also be positioned at 90° with respect to the detector.

ammonia confirming it as monochromatic THG radiation. Excitation spectra of the VUV emission exhibit increasing blue shifts and wider tuning ranges with increasing pressure, which is also shown in Fig. 3. Similar results have been recently reported for resonantly enhanced THG in xenon¹. Preliminary findings show that enhanced output may be possible by phase matching with a positively dispersive buffer gas.

Using higher CO pressures (> 200 Torr) and satisfying the phase matching requirements² will possibly allow continuous tuning over several hundred wavenumbers from the bandhead. Furthermore, if the higher $^1\Pi$ vibronic levels up to the predissociation limit also exhibit third-harmonic emission, tuning in the 1300-1500 Å wavelength region would result. However, for $v' \geq 3$ in the CO excited state manifold, the number of laser photons required to exceed the ionization potential changes from six to five allowing ionization to compete favorably with emission, thereby limiting the overall efficiency of the THG. The competition between ionization and emission is presently being interpreted by the two-level collective emission model³ put forth to analyze similar findings in xenon. The results of these studies will be discussed.

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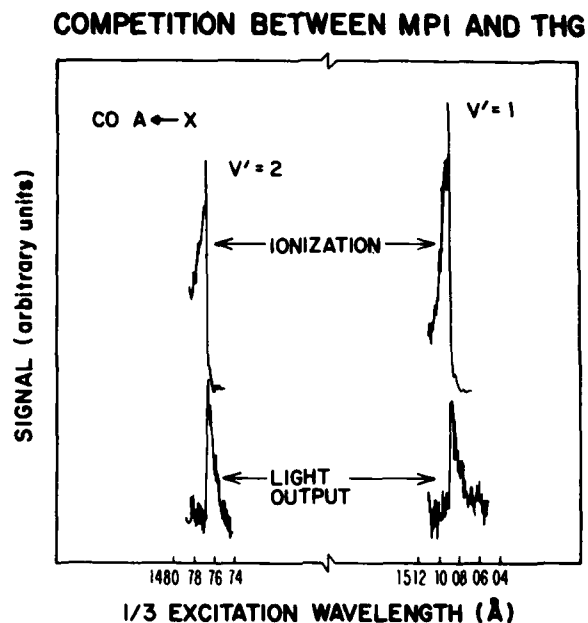


Fig. 2. The MPI and THG excitation spectra of the ${}^1\Pi$ $v' = 1$ and $v' = 2$ vibronic levels in CO at a pressure of 10 Torr. At higher dye laser dispersion the individual CO rotational lines are apparent in the MPI spectrum.

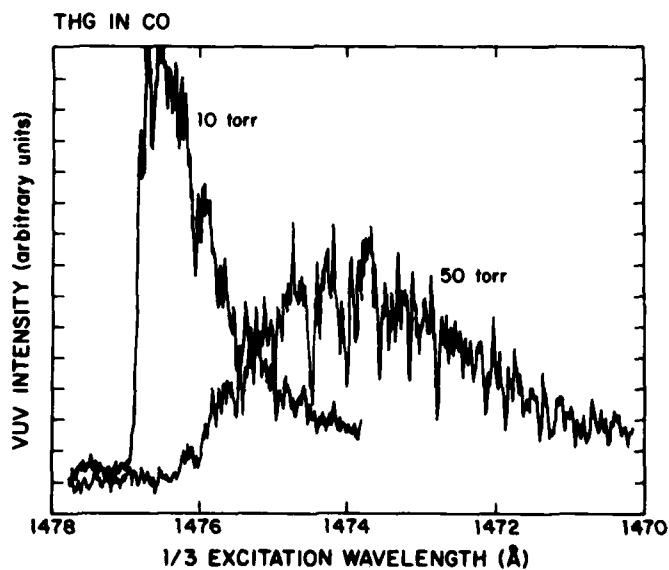


Fig. 3. Excitation spectra of the third-harmonic emission in the ${}^1\Pi$ $v' = 2$ level of CO at pressures of 10 and 50 Torr. The sharp dips in the VUV emission are due to absorption by rovibronic levels in the $\tilde{B} - 9$ band of ammonia.

Two-Photon Excitation of the Lowest g-States of Xe_2^*

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Applying two-photon excitation techniques we have studied atomic and molecular states in the energy region of the $5p^5 6s$ configuration of Xe I. In this way, states can be selectively populated which cannot be reached by dipole transitions from the ground state.

A detailed description of the experimental set-up is given in Ref. [1]. Frequency doubled laser light of 285-300 nm was focussed into a xenon gas vessel with typical power densities of 20 MW/cm^2 in the focus. The VUV fluorescence was detected using a solarblind photomultiplier and photon counting techniques. In Fig. 1, an example of the excitation spectrum is shown. The quadratic dependence of the fluorescence intensity on incident laser power and gas pressure prove that the observed structures arise from two-photon excitation of molecular states. In addition, the 3P_2 state was weakly excited, providing evidence for parity changing two-photon transitions.

From the results of polarization studies the symmetries of the excited molecular states were determined [2]. In view of theoretical potential energy curves [3] we arrive at an assignment for the molecular states as indicated in Fig. 1. Full consistency between theoretical and experimental data, however, could not be reached.

Information about the relaxation scheme of excited g-states can be obtained by spectral and time-resolved fluorescence studies. Fig. 2 summarizes first results. Discriminating the second continuum against the total fluorescence by means of a cutoff filter we observe different fluorescence behavior for g-states belonging to the 3P_1 or 3P_2 dissociation limits, indicating different

decay schemes. Further investigations, especially time-resolved studies of the fluorescence are in progress.

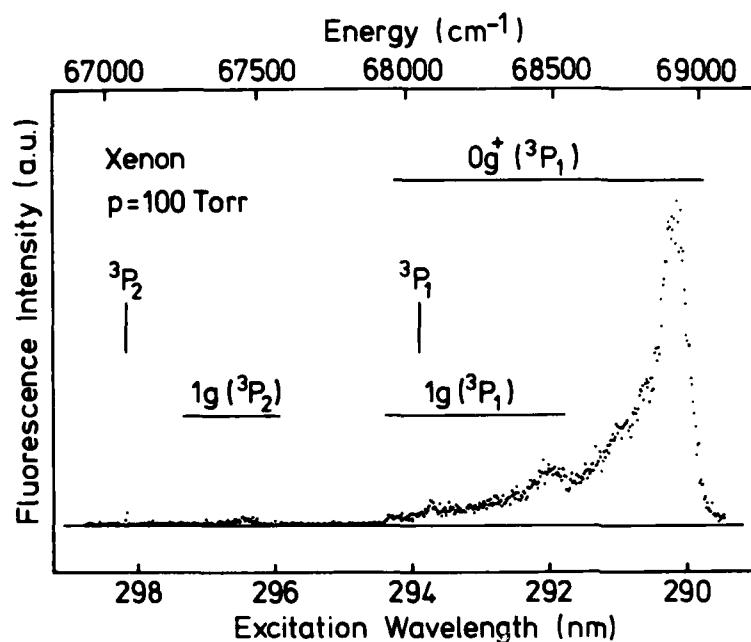


Fig. 1:

Two-photon excitation spectrum in the $5p^56s$ region of Xe I at 100 Torr gas pressure.

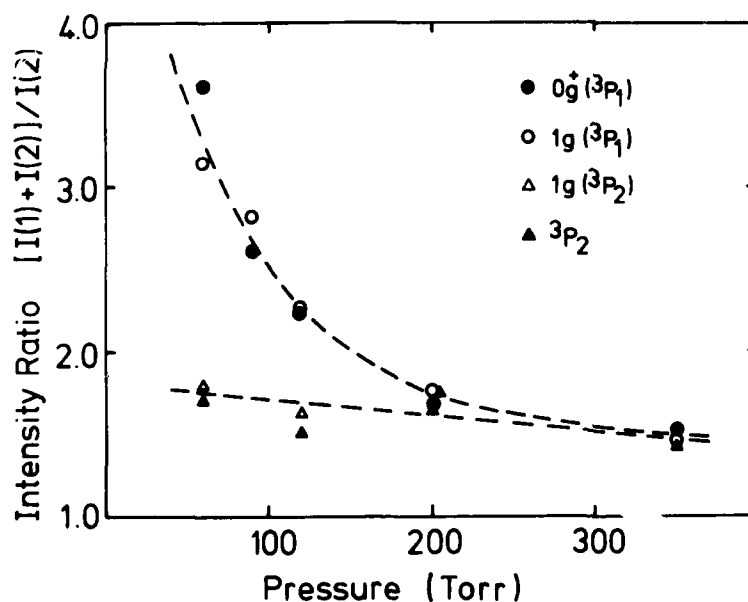


Fig. 2:

Fluorescence intensity ratios of the first I(1) and second I(2) continuum versus pressure. The intensities are not corrected for the spectral response of the detection system.

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Multiresonant Two-Photon-Absorption-Induced Four Wave Mixing in Crystalline
Rare Earth Insulators

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We report the first observation of intermediate state enhancement of four wave mixing induced by two photon absorption in rare earth insulators. Coherent generation of $\omega_4 = \omega_1 + \omega_2 - \omega_3$ exhibits strong intermediate state resonances when ω_1 sweeps through a single photon transition to the 5D_4 states arising from the $4f^8$ ground configuration of Tb^{3+} while $(\omega_1 + \omega_2)$ is simultaneously resonant with an allowed transition to the excited $4f^7 5d$ configuration. Measurements are presented for $Tb(OH)_3$ and $LiTbF_4$.

The coherent mixing experiments provide a number of advantages over conventional spectroscopic measurements including high resolution, new selection rules, and accessibility to new and relatively unexplored spectral regions using existing lasers. Significant potential exists for uncovering new laser transitions or materials and for elucidating the structure of excited configurations.

The nature of the process is confirmed by examination of power dependences and phase matching effects and by varying the temporal overlap of the three incident laser pulses. Coherent anti-Stokes Raman scattering is eliminated by choice of ω_2 and ω_3 .

Pulse sequenced experiments have demonstrated the expected free induction decay behavior of both the single photon (ω_1) and the two-photon ($\omega_1 + \omega_2$) excited states, proving the feasibility of using this method for picosecond coherent transient experiments. Our experiments have revealed substan-

tial line narrowing ($> 10X$) due to dispersive effects on the phase matching which may lead to selection of a homogeneous packet in coherent transient experiments on inhomogeneously broadened transitions. Present results, which we believe to be the first time domain measurements of coherence decay in a rare earth $4f^n-15d$ configuration, establish an upper limit of a few nanoseconds for the dephasing time. This technique is also applicable to rare earth vapor complexes where Doppler-free dephasing measurements are feasible.

Moderately strong laser powers of ten kilowatts focused to $100\text{ }\mu\text{m}$ spot diameters provide excellent signal to noise ratios at time constants of one second and 5 Hz repetition rates. Realistic potential exists for achieving "visible" output beams. Prospects for upconversion using $\omega_1 + \omega_2 \rightarrow \omega_3$ will be discussed.

Related results involving multistep excitation of fluorescence are presented, demonstrating major saturation effects in concentrated rare earth insulators at modest laser powers and focussing.

Research at Montana State University is supported in part by NSF grant # DMR 7906892 and 7918175.

Space resolved spectra of 2p-3d multiplets of Al XI and Al X....

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SUMMARY

The spectra of 2p-3d multiplets of Al XI and Al X emitted from plasmas produced from 9 μm thick Al foils were recorded with spatial resolution $\approx 30 \mu\text{m}$. The measurements are of interest in plasma diagnostics as well as in our search for suitable lasing transitions in the XUV {1}.

We used a conventional Nd: glass laser system delivering upto 10J energy in single pulses of about 180 ± 30 ps duration. A small pre-pulse of 20% nominal energy preceded the main laser pulse by 200 ps which was followed by an after-pulse of some 14% energy after another 200 ps. The laser was focused to $\approx 40 \mu\text{m}$ diameter spots and the spectra were recorded photographically using a 2 m grazing incidence spectrograph. The target foils were mounted on micromanipulators and we could position them accurately in the focal region such that the plasma expansion both in the forward (opposite to the laser beam) and the backward direction could be observed.

Fig. 1 shows the spectra of 2p-3d multiplets of Al XI recorded as a function of distance from the foil target. There are three lines in

the 2p-3d transitions of Al XI namely $^2P_{1/2} - ^2D_{3/2}$ (52.299 Å), $^2P_{3/2} - ^2P_{5/2}$ (52.446 Å) and $^2P_{3/2} - ^2D_{3/2}$ (52.442 Å) of which the last one is spectrally forbidden and this gives the transitions the shape of a doublet. For an optically thin plasma the doublet ratio should be $I_{52.299}/I_{52.446} \approx 0.50$ {2} as is the case if we integrate over the experimental line profiles for the plasma expanding in the forward direction (Fig. 1). However, the intensities recorded in the rear half space are measured to be in the ratio $I_{52.299}/I_{52.446} \approx 1.0$. In the case of Al X, the relative intensity ratios within the 2p-3d multiplet showed a similar departure from the expected behaviour.

The "anomaly" in the intensity behaviour may arise due to self-absorption in the relatively cooler outer layers of the plasma expanding in the rear of the foil. However, there is no evidence of self-reversal of line profiles of the type expected for optically thick plasma. This suggests that partial intermixing of quantum states may be occurring near the 3d states of Al XI and Al X {3}.

The laser heating and ionization in the plasma in this experiment were modelled assuming a self-similar expansion with a complete thermal burn-through of the foil {4}. A peak ionization of 10.4 was predicted by the model which is in satisfactory agreement with time averaged experimental observation of an average charge of 9.8 near the target surface on the front and 9.3 on the rear side. The slight overestimate of the average charge is ascribed to non-symmetric plasma expansion in the experiment and 100% absorption of laser light assumed in the code.

A two dimensional Eulerian hydrodynamic code {5} was also employed to simulate the multiple-pulse irradiation in this experiment.

Contrary to our experimental observation, the code did not predict hot plasma formation on the rear side of the foil. Experimentally, the role of pre-pulse (and possibly also of after-pulse) appears to be important. Our experiments with single pulse irradiation using comparable laser energy failed to produce observable plasma on the rear of the foil. The role of super thermal electrons which are not modelled in the Eulerian Code {5} may be significant here in producing the hot plasma at the rear of the foil. Due to the pre-pulse plasma, the main laser pulse interacts with a longer scale-length plasma. The production of superthermal electrons due to resonance absorption may decrease with longer scale-length plasma but it is likely to increase due to Raman scatter and two plasmon decay {6}. The superthermal electrons may preheat the target and aid the burn-through.

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Optical Density

0.20

0.15

0.10

0.05

0.00

52.446 Å

52.299 Å

52.446 Å

52.299 Å

52.299 Å + 52.446 Å (Unresolved)

52.446 Å

52.299 Å

300

200

100

0

-20

z (μm)

Resonant Four-wave Mixing Processes in Xenon

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Four-wave mixing processes have been widely used to generate coherent VUV radiation. In this paper we report the use of intermediate resonances in Xe to enhance the VUV output below 130 nm.

A dye laser, pumped by a doubled Nd:YAG laser, was doubled and mixed with the remaining IR beam ($\lambda_{\text{IR}} = 1.06 \mu\text{m}$) to produce a UV source tunable to Xe two-photon resonances. A $f = 500 \text{ mm}$ lens focused the UV and IR beams into a Xe cell. The generated VUV was detected by a solar-blind photomultiplier, after a monochromator and a VUV filter.

The generated down conversion signal ($\omega_3 = 2\omega_{\text{UV}} - \omega_{\text{IR}}$) was enhanced by more than a factor of 20 when the UV radiation was tuned through the two-photon resonant state $5p^5 ({}^2P_{1/2}^0) 7p [0\frac{1}{2}] J = 0$. Maximum signal resulted at a Xe pressure of about 25 torr. Saturation effects were absent at UV energies below 1 mJ and an IR energy near 700 μJ . Signals from a Xe ionization chamber have been interpreted as due to $\omega_4 = 2\omega_{\text{UV}} + \omega_{\text{IR}}$ at 101.7 nm. This wavelength could not be verified because of window absorption below 105 nm. Phase matching curves were obtained at both 125.9 nm and 101.7 nm; intensity dependences were observed and other intermediate resonances studied. Coherent radiation from other parametric processes was observed and relative strengths as a function of intermediate resonance detuning studied.

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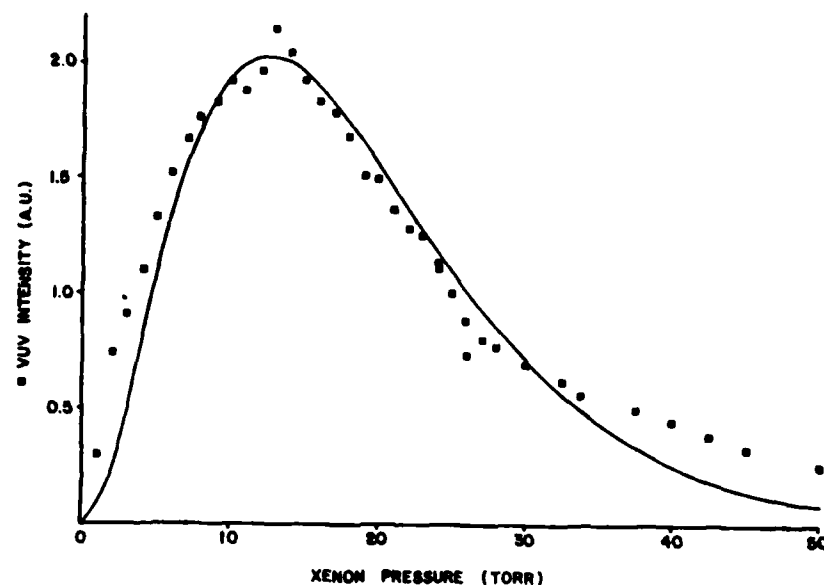


Fig. 1. Pressure dependence of VUV output: Experimental points \square and theoretical curve $— (\Lambda_1 \pi^2 P_2 e^{-\Lambda_2 P})$ for the down conversion signal ($\omega_3 = 2\omega_{UV} - \omega_{IR}$) due to the two-photon resonance state $7p[1\frac{1}{2}]_2$ of xenon.

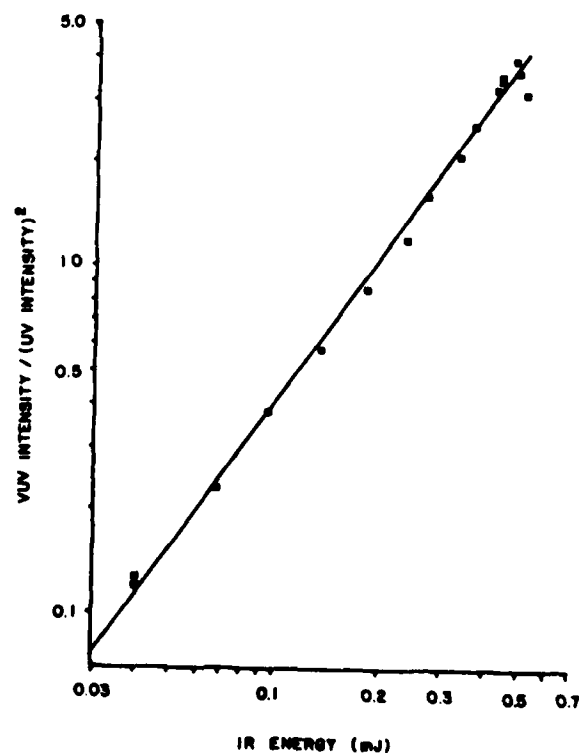


Fig. 2. I. R. intensity dependence of VUV output: Experimental measurements \square of the down conversion signal due to the resonance state $7p[1\frac{1}{2}]_0$ and the linear least-square fit $—$.

Vacuum UV laser induced fluorescence of the NO molecule

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Tunable narrow bandwidth VUV radiation in the spectral region from 190 - 200 nm was generated by resonant sum-frequency mixing of two flashlamp pumped dye lasers. This radiation was used to excite individual rotational vibrational levels of different electronic states of the NO molecule. Using a multiple reflection cell the effective quantum efficiency was raised and the subsequent fluorescence radiation was spectrally resolved by means of a 1m VUV grating monochromator. This technique allowed a direct measurement of the Franck-Condon factors for the different observed transitions. The measured values show significant discrepancies with calculations of Spindler et al./1/ neglecting the R-dependence of the electronic transition moment.

For the $A^2\Sigma(v'=3) - X^2\Pi(v''=1)$ transition the J-dependence of the transition probability was investigated. With increasing total angular momentum J the rotational splitting becomes comparable and even exceeds the multiplet splitting of the $^2\Pi$ ground state. This causes a change from Hund's coupling case a to Hund's coupling case b for increasing values of J.

Pumping the $C^2\Pi(v'=0)$ level, an additional fluorescence spectrum of the $A^2\Sigma(v'=0)$ level was observed. Since the

internuclear distances of the $C^2\Pi$ and $A^2\Sigma$ potential energy curves are almost identical, only transitions between levels with identical vibrational quantum numbers take place. As a result, a simple VUV fluorescence spectrum is obtained and the branching ratio for the $C^2\Pi - X^2\Pi$ and $C^2\Pi - A^2\Sigma$ transitions could be determined.

/1/ R.J. Spindler, Jr, L. Isaacson and T. Wentink, Jr
JQSRT 10, p.621 (1970)

Transform-Limited Bandwidth Injection Locking of an XeF Laser

Irving J. Bigio and Michael Slatkine

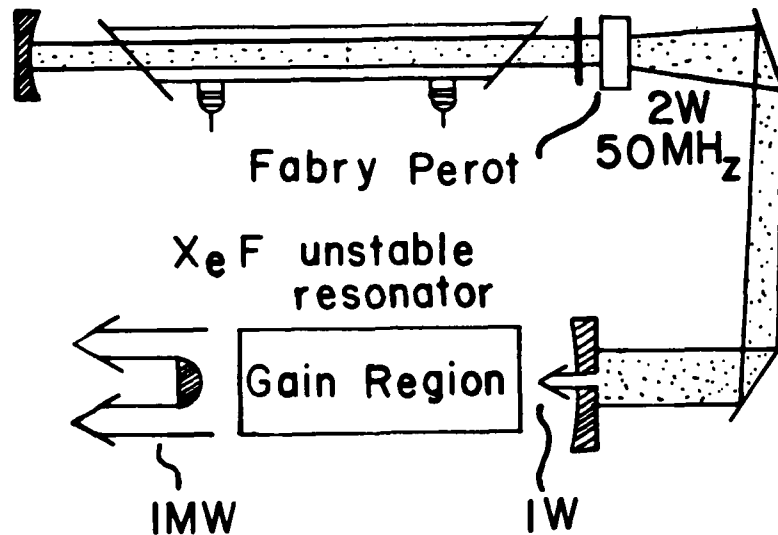
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Summary

Injection locking of excimer lasers has been shown¹ to be the most efficient way to control the spectral and spatial output of these high-gain systems. We have previously reported injection locking of a KrF system with very lower reference oscillator power, and in this paper we report the injection locking of an XeF laser to transform-limited bandwidth (~ 50 MHz for a 20-ns pulse). Injection locking of XeF has been previously reported,³ but the reference oscillator used was another XeF laser yielding a relatively broadband output linewidth of 0.2 \AA (~ 100 GHz).

The experimental setup is shown in the figure. The reference oscillator used here was a small and simple pulsed Ar-III ion laser operating on the 3511 \AA transition. The output was a 20-W, 0.5- μ s-duration pulse consisting of several longitudinal modes within the Doppler linewidth. A Fabry-Perot etalon was placed at the output to select one of the longitudinal modes, yielding a 2-W pulse of < 50 MHz bandwidth. Approximately 1 W was then injected through a small hole in the rear optic of a positive-branch confocal unstable resonator. The XeF gain medium was a discharge pumped volume $2 \times 2 \times 60 \text{ cm}$.

Ar Ion Laser, 3511 Å°



The total laser output was ~ 50 mJ in 25 ns and was derived from many vibrational-rotational lines in the regions of 349, 351, and 353 nm. Because the output originates from several different upper levels, not all of the energy is available to be locked onto the reference oscillator wavelength. Our measurements show that about 60% of the total output can be channeled into the 50 MHz bandwidth at the locking frequency. This can serve as an indirect measurement of the communication rate among the upper levels. The laser beam divergence was also measured to be near diffraction limited.

We have recently learned that similar results have been achieved with KrF^4 by injection locking with the frequency-doubled 496 nm line of an Ar-II ion laser. These injection locking techniques demonstrate that ultra high spectral brightness is available from

excimer lasers with relatively uncomplicated and moderate-cost systems, enabling many more researchers to probe nonlinear optics in the uv. Indeed we have already used these systems to study backward stimulated Brillouin and Raman scattering as well as four-wave mixing.

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Excitation and Ionization of Atomic Hydrogen by
Narrow Bandwidth Tunable VUV Laser Radiation

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By frequency mixing in rare gases it is now possible to generate tunable coherent radiation in the entire wavelength region from 105 nm to 200 nm¹. This techniques will thus open the way for many applications of tunable VUV radiation to physical and photochemical problems. In this contribution we want to report on results of the excitation of atomic hydrogen in an atomic beam. The tunable VUV radiation excited the $1^2S_{1/2} \rightarrow 2^2P$ Lyman - α transition, and a second tunable UV laser served to excite the $(2^2P) \rightarrow H$ atoms further to high Rydberg states or the ionization continuum. Preliminary results together with a detailed description of the apparatus has been published^{2,3}.

For the generation of narrow bandwidth tunable VUV radiation by frequency mixing in atomic gases the fundamental radiation, in this case at $\lambda = 364$ nm, has also to be narrow in bandwidth. We were using nearly Fourier transform limited tunable dye laser radiation around $\lambda = 554.5$ nm ($\Delta\nu_0 \approx 100$ MHz) as the main tunable source⁴. This radiation was mixed with the IR output of a Nd:YAG laser at $\lambda = 1064$ nm in a KD*P crystal, yielding efficient generation of near UV light around $\lambda \approx 364$ nm. The bandwidth of the Nd:YAG laser was narrowed by using an electronically controlled opening of the Q-switch together with two intracavity etalons, a 3.5 mm air spaced one (finesse ≈ 15) and a 13 mm fused quartz one with finesse ≈ 5.5 . We obtained

an operation of the Nd:YAG laser with Fourier transform limited bandwidth ($\Delta\nu \approx 60$ MHz). Because the laser resonator was not stabilized, the frequency jittered between two longitudinal modes, separated about 250 MHz. Focussing the near UV radiation ($\lambda = 364$ nm) with a quartz lens ($f = 70$ mm) into a cell containing krypton gas ($p = 130$ mbar) yielded about 5 nJ/pulse linearly polarized VUV radiation with a bandwidth of about 500 - 700 MHz, monitored over several thousands of pulses. Circularly polarized VUV light was obtained by inserting a MgF_2 quarter wave plate in the beam.

The tunable VUV radiation excited the $(1s^2S_{1/2}) \rightarrow (2p^2P)$ Lyman - α transition in an atomic hydrogen beam. Due to the narrow bandwidth of the VUV laser the ground state hyperfine structure splitting could be resolved. Using circularly polarized light and starting from the ($F = 0$) hyperfine state in $1s^2S_{1/2}$ it is possible to populate a single m_F state in $2p^2P_{1/2}$, thus polarizing the hydrogen atom in the excited state. Results on photoionization of such polarized atoms will be reported.

In a different experiment the VUV radiation was separated from the generating near UV light by a MgF_2 prism. The VUV laser beam was then directed to cross an atomic hydrogen beam in a region, where all nearby surfaces were held at a temperature of 14 K. A second anti-collinear tunable UV laser ($\lambda = 360 - 375$ nm) served to further excite the $(2p^2P)$ - H atoms into high Rydberg states ($n \approx 50$). The excitation of Rydberg states was detected by field ionization. Results on the field ionization of S and D Rydberg states will be reported.

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PLASMAS I

Review of Upconverted Nd-Glass Laser Plasma Experiments
at the Lawrence Livermore National Laboratory*

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Abstract

Much has been learned about laser heated disk target plasmas which may find application in diverse technologies.

Summary

Systematic experiments aimed at deducing the dependence of laser-plasma interaction phenomena on target plasma material and target irradiation history have been underway in laboratories all over the world in recent years. These measurements have provided data useful in many optical technologies. During 1980 and 1981 the Livermore program undertook to measure the laser light absorption of high and low Z disk target plasmas and the partition of the absorbed energy amongst the thermal and suprathermal electron populations as a function of both laser intensity and wavelength. Simulations suggested that short wavelength laser light would couple more efficiently than longer wavelengths to target plasmas. Shorter wavelength heating of higher electron plasma densities would, it was felt, lead to laser-plasma interactions freer of "anomalous" absorption processes. The experiments to be reviewed were designed to test these hypotheses.

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Resonantly-Pumped Soft X-ray Lasers Using ICF Drivers

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Simulations of soft X-ray laser targets irradiated with 100 psec pulse of 1.06μ light at $2-4 \times 10^{14}$ W/cm² indicate that small signal laser gain between 10 and 50 cm⁻¹ may be obtained on 4-3 transitions in helium-like F and Ne (42.3 eV and 53.6 eV) as well as on 3-2 transitions in helium-like and hydrogenic F and Ne (119 eV-153 eV). Resonances include the Na X/Ne IX resonance near 11.000 Å as well as several L-shell/K-shell resonances (for example, Be-like Cr (13.779 Å)/He-like F (13.782 Å)).

The laser target consists of flashlamp and filter systems surrounding the laser medium (which occurs as a gas). Irradiation of the flashlamp causes X-ray production which transiently strips the laser medium to the appropriate (He-like, H-like) sequence. Strong line emission may then resonantly pump an inversion if the transverse gas dimensions are near 50μ. In the case of 3-2 lasers, radiation trapping is important, and we have used a 2-D angle- and time-dependent PRD model for the simulations.

TuA3-1

Generation of Vacuum Ultraviolet
Radiation by Plasma Nonlinearities

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Summary:

Early experimental work¹ and numerical studies² have examined the production of more than fifty harmonics (212 nm) of the CO₂ laser in laser matter interaction studies carried out at Los Alamos within the laser fusion program. Initially it was believed that pressure balance between hydrodynamic plasma expansion and radiation pressure was responsible for a substantial density jump near critical, leading to the limitation on the number of harmonics produced.² While still believed to be important, more recent CO₂ laser studies³ have revealed that early-time hydrodynamics when combined with the laser skin depth can play a very significant role in the production of high harmonics for CO₂ laser matter interaction experiments conducted at high irradiances.

In this paper, we will present the results of detailed hydrodynamics studies and analytic models which explain a large range of CO₂ laser harmonic experiments conducted within the laser fusion program. We then extend these calculations to the case of a high power ArF laser matter experiment where it is predicted that about seven (27.6 nm) to ten (19.3 nm) harmonics should be observed at high conversion efficiency ($\sim 10^{-4}$ /harmonics). The details of these calculations and their range of applicability will be discussed.

1. R. L. Carman, C. K. Rhodes, and R. F. Benjamin, Phys. Rev. A 24, Nov. 1981.
2. R. L. Carman, D. W. Forslund, and J. M. Kindel, Phys. Rev. Lett. 46, 29 (1981).
3. R. L. Carman, Proc. Topical Conf. on Symmetry Aspects of Inertial Fusion Implosions, Ed. Naval Research Laboratory, 1981.

A SOFT X-RAY SOURCE BASED ON THE MAGNETIC CONFINEMENT AND
COMPRESSION OF A LASER PRODUCED PLASMA

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Summary

The physics of laser heating of dense plasmas produced by irradiation of solid targets, and of underdense ($N_e < \text{critical density}$) plasmas has attracted considerable interest in recent years. Experiments at MIT¹ and at MSNW have recently demonstrated radial magnetic confinement of a solid target plasma plume produced by radiation from a 10.6μ CO₂ laser. Presently generated plumes of moderate Z ions are at 100 eV temperatures, 10^{18} cm⁻³ densities, and last about 100 nsec. The addition of a simple z-pinch could compress the plume to 10^{19} cm⁻³ densities and 600 eV temperatures where it would be an extremely efficient generator of K or L-shell x-rays in the 1-2 keV range. The efficiency and simplicity of such a device, plus the proven physics of the laser plume generation hold great promise for commercial microlithography applications.

The MSNW experiments were done using solid carbon targets which were irradiated with CO₂ laser pulses of up to 500 J and 50 nsec (FWHM) duration. The targets were immersed in an axial magnetic field of 7.5 T. The resulting plasma is confined radially and expands as a plume toward the heating laser.

Plasma conditions in the plume were investigated using ruby laser holographic interferometry to determine electron density profiles and spatially resolved Thomson scattering to determine electron temperature profiles. The resulting data was found to be in good agreement with a high beta pressure balance plasma model. Typical plasma conditions in the plume are $T_e = 100\text{--}150\text{ eV}$, $N_e = 1.0 - 1.2 \times 10^{18}\text{ cm}^{-3}$. The electron temperature is extremely uniform due, in part, to radial heat conduction, but primarily to the strong temperature dependence of the laser absorption coefficient.

We have closely coupled our experimental programs with computer modeling of present and contemplated experiments, using various MHD codes with a collisional-radiative atomic physics package. A one-dimensional radial code, DYNASOR, successfully predicted the observed radial profiles in the solid target experiments, while a one-dimensional, axial code, CORK, has given reasonable agreement with the axial structure of the plume. We will present results of these plume simulations. In addition, we will present results of simulations of the z-pinch compression using an extended DYNASOR code. Implications of these results to the feasibility of a commercial x-ray source based on this principle will be discussed.

1. Interaction of CO_2 Laser Pulses with Solid Targets in Magnetic Fields, N.G. Loter, W. Halvorson, and B. Lax, J. Appl. Phys. 52, (8) August 1981 (P5014).

PLASMAS II

A Laser Produced Plasma as an Extreme Ultraviolet Continuum Source for absorption Experiments

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For experiment of absorption of highly ionized species, produced in a pulsed regime, a background source emitting continuum in the XUV region with high brightness is needed. The spectral output of plasmas produced by focusing a ruby laser pulse of 10 J of energy and 15 ns duration on plane targets of various light materials has been studied.

The region of highest brightness of the plasma is the ablation region where the density approaches the critical density ($n_e = 2.2 \times 10^{21} \text{ cm}^{-3}$ for ruby). On plane targets such region stays below the unperturbed surface of the material and consequently a deflecting and focusing system that observes the plasma end-on, as shown in Fig. 1, was used. The laser beam is deflected by the mirror A and focused by the lens L_1 on target T_1 . The XUV radiation is observed through holes in both A and L_1 and focused by the toroidal mirror M working at grazing incidence on the entrance slit S of a grazing incidence spectrometer. Other advantages of the system used are the complete filling of the aperture of the spectrograph and the nearly total compensation of the astigmatism of the spherical grating G.

With a grating having 2400 lines/mm, a 2m radius of curvature and used at an angle of incidence of 86° and a 5 μm wide entrance slit good spectra were recorded with single shots on Kodak 101-01 films.

The instrumental spectral broadening was $\approx 10^{-2} \text{ \AA}$. The system was absolutely calibrated both with a branching ratio method and by measuring the efficiency of the grating. The spectrum emitted by a carbon plasma in the region 20-50 \AA is shown in Fig. 2. The spectrum consists of a free-free and free-bound continua and few resonance lines of CV and CVI. With dotted lines the continuum spectrum predicted by a plasma model is shown [1]. Changing the material of the target the emitted spectrum can be shifted to shorter or longer regions. By using materials with high atomic number (e.g. Cu) a

nearly line-free continuum is emitted. The brightness of such laser produced plasmas compares favorably with the synchrotron radiation in the same spectral range. By integrating the emitted spectra over wavelength, solid angle and size of the source a total amount of power of ≈ 4 MW is obtained; this represent about 1% of the laser energy absorbed by the target. Ways of improving the output of the plasma have been studied like firing the laser on a previously built crater and using long focal length lenses. The laser produced plasma can be considered a reliable and useful XUV source for fundamental investigations of atoms and ions. The present source is used on an experiment of absorption spectroscopy of ions with high spectral resolution in the XUV. A second laser produced plasma optimized for ions production is produced as shown in Fig. 1 by lens L_2 on target T_2 near the entrance slit S of the spectrograph. The region of absorption on the expanding plasma can be moved up and down and the time delay between the two plasmas, the source and the absorbing one, varied. The velocity spreading on the expanding plasma [2] has been reduced to the equivalent of $\approx 5 \times 10^{-3}$ Å at 150 Å by using a suitable contribution of focusing lens and a preformed cavity on the target T_2 .

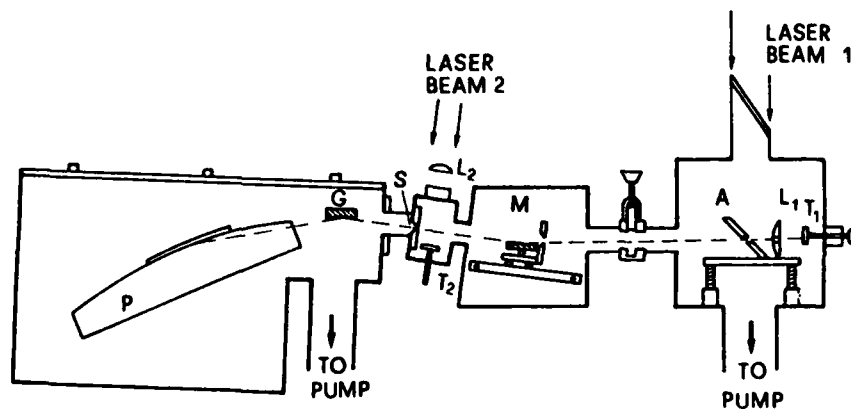


fig. 1

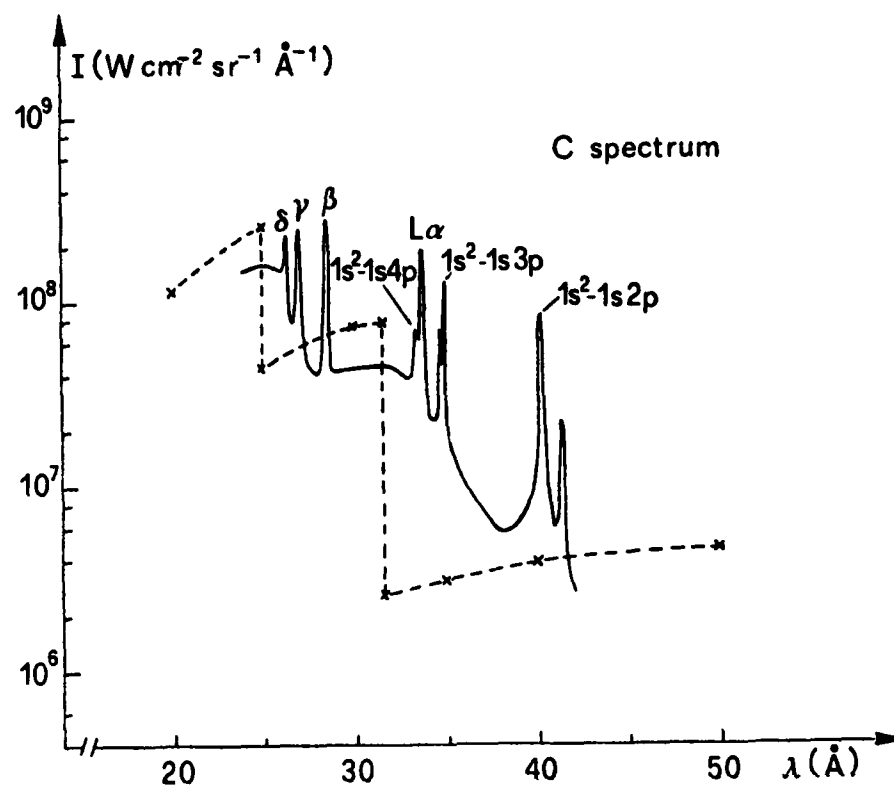


fig. 2

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- [2] E. Jannitti, P. Nicolosi and G. Tondello, 15th European Conference on Laser Interaction with Matter, (1982).

Optical Techniques for Use with Soft X-ray and XUV Radiation

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SUMMARY

The application of x-ray and XUV radiation to a wide range of interesting biological and physical phenomena will require not only sources of radiation, but also the availability of appropriate "optical" components and techniques for manipulation, transport and detection. Recent technological advances, driven by widely differing scientific requirements, primarily in the areas of micro-electronics and hot, dense plasma studies, have led to broad based and rapidly emerging capabilities in this area. The fabrication of structures with characteristic dimensions on the scale of soft x-ray wavelengths (10 to 100's Å) is now possible. These diffractive elements are opening new opportunities with laboratory and astrophysical x-ray sources, as x-ray interference mirrors and splitters, x-ray lenses, transmission gratings and microscopes. Their use with quasi-coherent radiation sources in x-ray interference and holographic experiments will provide exciting new possibilities. In addition, parallel efforts have led to improved detectors, including picosecond soft x-ray streak cameras.

These emerging technologies represent the work of contributors of widely differing interests at universities, national laboratories and industry. The breadth of these developments in x-ray optics has only

recently been brought to the attention of the major contributors themselves.¹ It is our purpose in this conference presentation to review and reference the more significant developments, in timely fashion, for the related community involved in the development of XUV sources of radiation.

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Population Density and VUV Gain Measurements in Laser-Produced Plasmas

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Population density inversions in laser-produced plasmas between hydrogenic and helium-like carbon ion energy levels were obtained from measured resonance line comparisons^{1,2}. Such measurements have been extended to absolute flux from which the densities of potential lasing transitions are deduced. From this and the line width the gain coefficient can be derived. This becomes a vital gauge diagnostic in increasing the gain towards significant amplification by parameter variation for increased ion density and increased pumping. Calibration of the vacuum spectrograph for absolute intensity determinations is performed in the near-uv against a standard D₂ lamp and transferred to the 4 nm wavelength region by an in-situ branching ratio technique involving resonance as well as high-lying transitions. Gains measured in electron-capture pumped plasmas are in agreement with a simple physical model from which parameter scaling is readily conceptualized. Expansion of the parameter space available for significant gain is predicted for higher-Z targets and for supplementary photon pumping.

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²R. C. Elton, T. N. Lee, R. H. Dixon, J. D. Hedden, and J. F. Seely, in Laser Interactions and Related Plasma Phenomena, H. J. Schwarz, H. Hora, M. Lubin and B. Yaakobi, editors (Plenum Publ. Co., 1981).

Pulsed Hollow Cathode Discharge for XUV Lasers
and Radiation Sources

by

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Recent proposals for short wavelength ($< 1000 \text{ \AA}$) lasers¹ have indicated a need for new types of electric discharge devices which produce significant densities of highly excited atoms and ions. In particular, the proposal of Harris¹ for a laser at 200 \AA using the doubly excited states of lithium requires the production of excited atoms in states with $\sim 60 \text{ eV}$ energy at densities $\geq 10^{12} \text{ atoms/cm}^3$. This Letter reports the operation of a pulsed hollow cathode discharge (HCD) which creates population densities of $4 \times 10^{12} \text{ ions/cm}^3$ in the lithium ion $1s2s \text{ }^3\text{S}$ level at 59 eV above the ion ground state, $\text{Li}^+(1s^2 \text{ }^1\text{S})$. Population densities of other relevant levels as well as electrical characteristics of the HCD have been measured.

The discharge operates as a heat pipe (to obtain high ground state metal vapor densities) and at high current density (25 amps/cm^2) and high voltage (1000 V) to achieve appreciable excitation rates of highly excited levels.

A summary of our population measurements is given in Table 1. These measurements were made at the peak of the highly excited state population densities which occurred $\sim 200 \text{ ns}$ after the start of the current pulse. These results may be useful for the realization of proposed schemes for discharge excited XUV lasers¹ and for short wavelength light sources based on anti-Stokes Raman scattering from highly excited metastable states.²

| Species | Number Density (atoms/cm ³) |
|---------------------------------------|---|
| Li(1s ² 2s) | 2×10^{16} |
| Li(1s ² 2p) | 8×10^{15} |
| Li ⁺ (1s ²) | $< 1 \times 10^{14}$ |
| Li ⁺ (1s2s ³ S) | 4×10^{12} |
| Li ⁺ (1s2s ¹ S) | 5×10^{11} |

Table 1

Lithium Population Densities Measured in HCD

References

1. S. E. Harris, R. W. Falcone, M. Gross, R. Normandin, K. D. Pedrotti, J. E. Rothenberg, J. C. Wang, J. R. Willison, and J. F. Young, "Anti-Stokes Scattering as an XUV Radiation Source," Paper No. 78 presented at the Fifth International Conference on Laser Spectroscopy, Jasper, Canada, June 29-July 3, 1981.
2. J. E. Rothenberg, J. F. Young, and S. E. Harris, Opt. Lett. 6, 363 (1981).

MULTIPHOTON PROCESSES I

Multiphoton Ionization of Atoms and Molecules

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Recent studies of resonantly enhanced multiphoton ionization (MPI) and third-harmonic generation (THG) in the rare gases Xe, Kr, and Ar, will be discussed. In particular, the disappearance with increasing pressure of the MPI signals resonant with the three-photon allowed 6s, 5s, and 4s states, respectively, of the rare gases is correlated with the appearance of THG. At certain laser wavelengths the THG signal is partially quenched by self absorption.

Measurements of the electron and fragment ion kinetic energy distributions resulting from resonantly enhanced MPI of molecules is adding considerable new insight into the basic physics of such processes. High resolution (70 meV FWHM) multiphoton photoelectron kinetic energy spectra for NO, H₂S, NH₃, and C₆H₆ will be presented and discussed in relation to the primary ionization event and subsequent photofragmentation processes.

¹Operated by Union Carbide Corporation with the U.S. Department of Energy.

First Observation of Photoionization of Laser-Excited Atoms by Synchrotron Radiation (SR)*. J.M. BIZAU,F. WUILLEUMIER, P. DHEZ, Lab. Spect. Atom. et Ion. and
LURE, Orsay; D. EDERER, NBS, Washington, DC; J.L. LEGOUET,
J.L. PICQUE, Lab. Aimé Cotton, Orsay; P. KOCH, Yale Univ.

New Haven, CT.*--In a triple, orthogonal crossed beam experiment, we have studied photoionization of excited Na atoms. A cw ring dye laser (few W/cm^2) locked to the D_1 or D_2 absorption lines produced a steady-state fraction up to 20% of excited $\text{Na}(3p^2P_{1/2}$ or $3p^2P_{3/2})$ atoms in a weakly collimated $\text{Na}(3s^2S_{1/2})$ beam with density $\lesssim 10^{13} \text{ cm}^{-3}$. Monochromatized SR from the ACO storage ring provided the photoionizing radiation. A cylindrical mirror electron spectrometer was used to measure photoelectron spectra. First measurements of photoionization from the 2p subshell with the 3p orbital occupied were obtained. Transitions to autoionizing resonances $h\nu_{\text{SR}} + \text{Na}(2p^6 3p^2 P_J) \rightarrow \text{Na}(2p^5 3s 3p)^1$ were driven near $h\nu_{\text{SR}} \sim 31 \text{ eV}$ from either $J=1/2$ or $J=3/2$ levels. A comparison of the results for the $2p^6 3p \rightarrow 2p^5 3p \epsilon d$ and $2p^6 3s \rightarrow 2p^5 3s \epsilon l'$ ionization channels will be given.

¹J. Sugar et al., Opt. Lett. 4, 109 (1979).

*Work carried out at LURE, Univ. Paris Sud, Orsay, FRANCE.

Submitted by

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A251 Physics
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Electron Spectra of Four-Photon Ionization of NO

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We are currently engaged in a program on spectroscopy of electrons created in multi-photon ionization. Results on atomic Xe concerning additional absorption of photons in the N-photon continuum have been reported recently [1]. These data were obtained using a novel type of electron spectrometer described in a separate abstract.

We now present results on four-photon ionization of NO using the same technique. We record electron spectra at various wavelengths within the rotational width of the two-photon intermediate resonance with the $A^2\Sigma^+(\nu=1)$ state. In a similar experiment [2] only two electron energies were observed, at 2 eV near zero. Our instrument permits full resolution of the spectrum and produces peaks corresponding to formation of NO^+ ions in each of the energetically allowed vibrational states. The intensity distribution deviates strongly from that of a smooth Franck-Condon envelope, which we ascribe to autoionization. Complications of the spectra arising from additional absorption in the continuum or absorption from long-lived excited states can be resolved by accurately (< 2 meV) measuring the shift in peak energy with wavelength.

[1] - P. Kruit, J. Kimman and M.J. Van der Wiel,

J. Phys. B. 14 (1981) L 597

[2] - J. Miller and R.N. Compton

J. Chem. Phys. 75 (1981) 22

-- The authors have no preference for oral or poster presentation. --

GENERATION OF NARROW-BAND TUNABLE RADIATION IN THE 1200A

SPECTRAL REGION

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In an extension of previous work^{1,2} on the efficient (up to 1%) generation of vacuum ultraviolet radiation in mercury vapor, we now report the generation of continuously tuneable, narrow-band (0.02 cm^{-1}) radiation in the spectral region of 1218Å to 1197Å. The mechanism is a resonantly enhanced 4-wave sum-mixing process $6s^2 1S_0 \xrightarrow{\omega_1} 6s7s^1 S_0 \xrightarrow{\omega_2} 6snp^1 P_1 \xrightarrow{\omega_3} 6s^2 1S_0$ where the ω_3 radiation is generated about the 12p through the $16p^1 P_1$ transitions of Hg. The ω_2 coumarin 520 dye laser is pumped by the third harmonic of a Nd:Yag laser, currently producing only 0.2 mJ of laser light which is tuneable from 5000 to 5500Å. The ω_1 laser at 6250Å is pumped by the remaining second harmonic of the Nd:Yag giving 0.8 mJ when frequency doubled to 3125Å. We are confident that these input energies will be increased substantially when the appropriately coated optics are acquired. Using a 25 cm focal length achromat to focus the beams into a Hg cell at 140°C (2 Torr), with 10 Torr of helium as buffer gas, we measure the generated photons in an NO ionisation chamber. The generated power peaks up substantially in the immediate vicinity of the $np^1 P_1$ transition frequencies and, even though hydrogen Lyman α is almost exactly midway between the 11p and the 12p transition, we measure 2×10^9 photons per pulse in this 10 pps system. At the nearby 12p $1P_1$ frequency (1212.65Å) we see 2×10^{11} photons per pulse. We will show absorption profiles scanned through hydrogen Ly α (1215.67Å), deuterium Ly α (1215.34Å) and also through the resonance

triplet of atomic nitrogen at 1199.55Å, 1200.22Å, and 1200.71Å. A careful examination of the wavelengths involved lead to the conclusion that the transition line frequencies listed in the Moore tables for the 15p and the 16p¹P₁ levels of Hg are off. We measure the 15p level to be at 83273.5 cm⁻¹ whereas the Moore tables give 83280.5 cm⁻¹. Similarly, the 16p is measured at 83407.7 cm⁻¹ in comparison with the tabulated frequency of 83420.3 cm⁻¹.

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MULTIPHOTON PROCESSES II

Multistep Excitation of Alkaline Earth Atoms to Autoionizing States

T. F. Gallagher

Molecular Physics Laboratory

SRI International

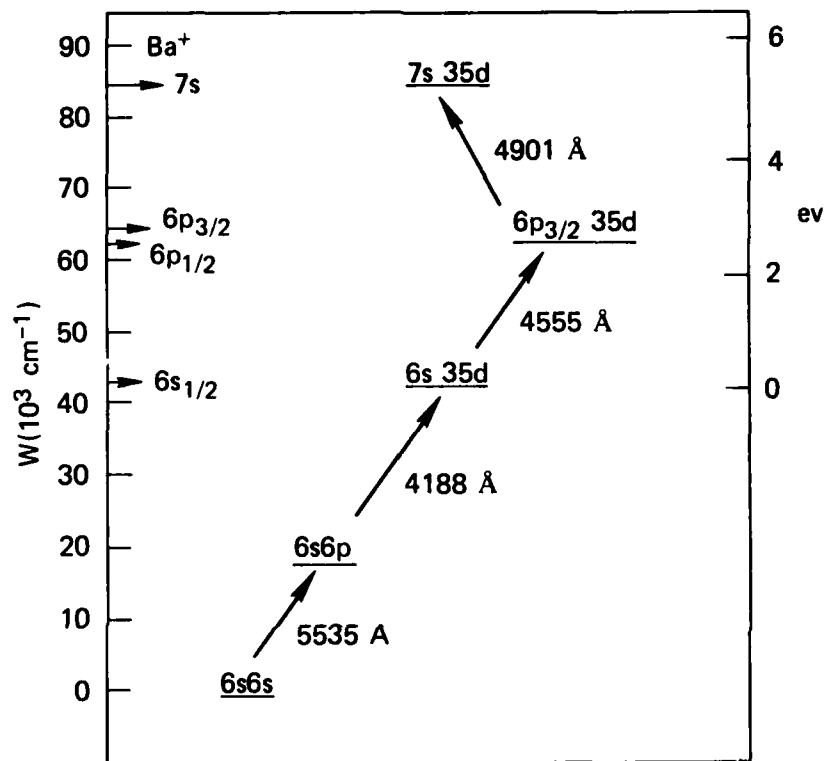
Menlo Park, CA 94025

Summary

Using a multistep laser excitation of alkaline earth atoms in an atomic beam we are able to excite autoionizing states in which one of the electrons can be in a state of almost arbitrary n and l . The multistep approach uses all single photon transitions in which one electron is excited at a time, as shown in Fig. 1. In addition to the technical simplification of requiring low energy photons, exciting one electron at a time simplifies the observed spectra and their interpretation.

Since these experiments are done in a low density atomic beam we have added an electrostatic electron energy analyzer which enables us to detect the final states of the ion and electron following autoionization.

The excitation scheme, and its application to the optical spectroscopy, and electron spectroscopy of alkaline earth autoionizing states will be discussed.



SA-3456-109

Fig. 1. Ba energy levels for the excitation of the 7s nd states

Vacuum Ultraviolet Spectroscopy of Molecules Using Third-Harmonic Generation in Rare Gases

John C. Miller, C. D. Cooper, and R. N. Compton

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We describe a simple apparatus for spectroscopic studies in the vacuum ultraviolet (VUV) spectral region which is based on third-harmonic generation (THG) in rare gases. The apparatus, shown in Fig. 1, consists of three chambers, each of which functions as a proportional counter. Light from an N_2 -pumped dye laser is focused into the first chamber containing the tripling gas. The VUV radiation is then collimated and passed through a sample cell containing flat-plate electrodes and into the third chamber. This last cell acts as a VUV photon detector via the photoelectric effect. In all three chambers electrons, produced by ionization, are amplified in a counter gas and detected at the electrodes.

We have demonstrated the utility and simplicity of this VUV spectrometer by performing a variety of spectroscopic studies. Data will be presented illustrating one-photon absorption spectra of states of CO below the ionization potential (IP) and one-photon photoionization studies of CH_3I which probe autoionizing states. The IP of iodobenzene has been determined by a threshold study to be 8.75 eV.

As THG results in colinear beams of VUV and blue light, we can also perform two-photon, two-color experiments. For instance, the VUV light has been used to excite NO into the $F^2_{\Delta}(v=3)$ or $N^2_{\Delta}(v=1)$ Rydberg states while the blue light subsequently ionized the NO as shown in Fig. 2.

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TOPICAL MEETING ON LASER TECHNIQUES FOR EXTREME
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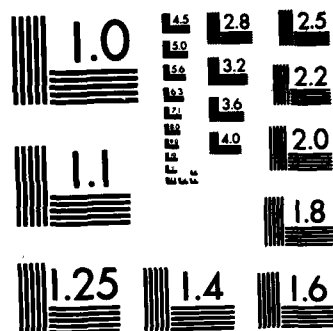
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Vacuum Ultraviolet Spectroscopy of Molecules Using Third-Harmonic Generation in Rare Gases

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Detailed comparison of the absorption spectrum in Fig. 3b with the two-photon ionization spectra of Fig. 3a shows that some simplification occurs in the two-color experiment. States which rapidly predissociate do not appear in the two-photon ionization thus simplifying regions which are congested in the absorption spectra.

¹Operated by Union Carbide Corporation with the U.S. Department of Energy.

ABSORPTION AND IONIZATION APPARATUS USING THIRD-HARMONIC GENERATION

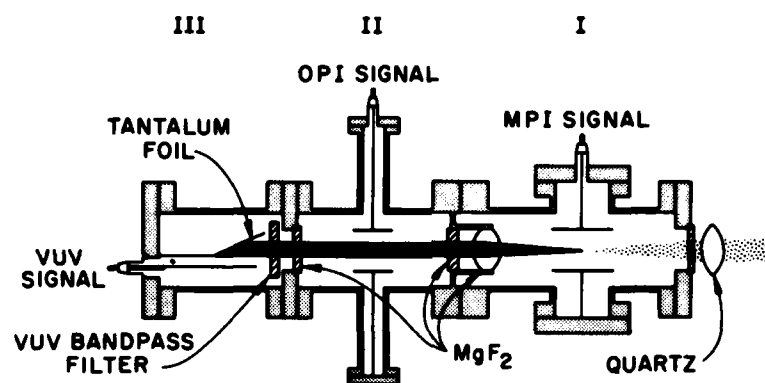


Fig. 1

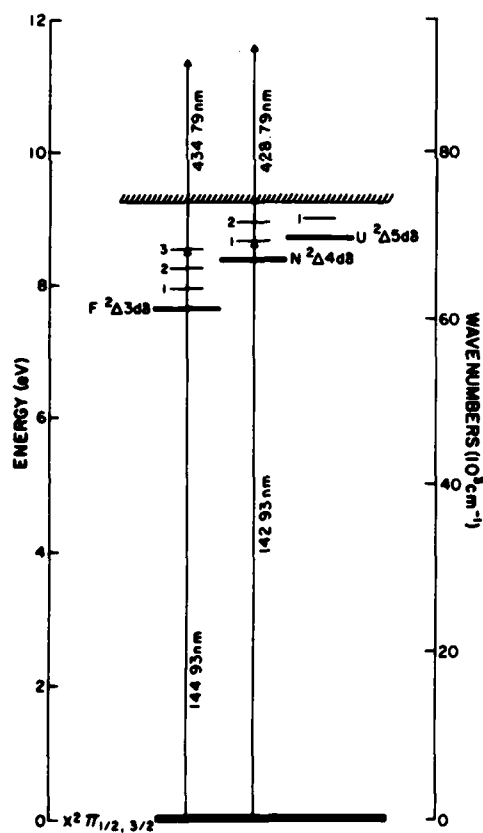


Fig. 2

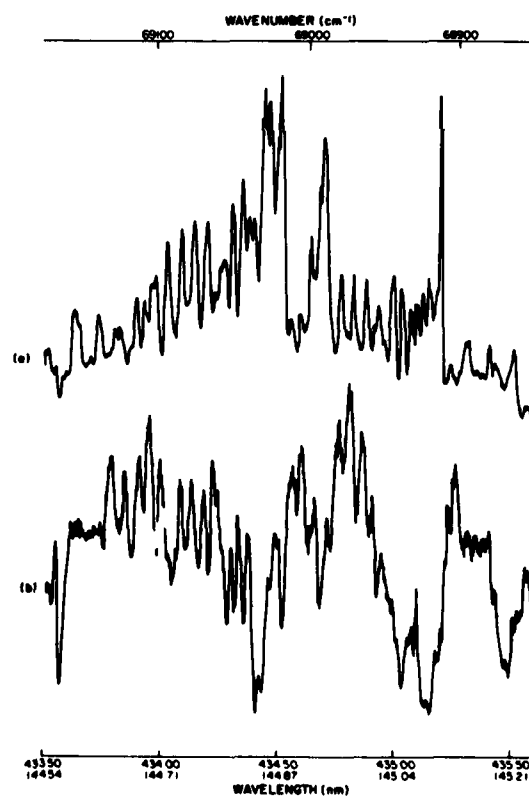


Fig. 3

NON-LINEAR MIXING I

NON-LINEAR MIXING

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Summary

We report investigations with tunable VUV radiation, employing VUV generation in regions down to the cut-off wavelength of lithium fluorid crystal (~ 110 nm) by frequency tripling of tunable UV from dye laser. - Experiments on multiphoton excitation and ionization of atomic hydrogen with tunable VUV light at Lyman- α , have been carried out with the following objectives: (1) sensitive H and D atom detection by two-photon ionization i. e. $1s \xrightarrow{\omega_{\text{Ly}\alpha}} 2p \xrightarrow{\omega_{\text{UV}}} \text{ion}$, (2) two-photon ionization of H with high resolution at Lyman- α to resolve the hyperfine groundstate doublet, aiming at the production of polarized protons, (3) state selective production of high Rydberg states ($1s \xrightarrow{\omega_{\text{Ly}\alpha}} 2p \xrightarrow{\omega_{\text{UV}}} ns, nd$), and their ionization, (4) Doppler line shape spectroscopy at Lyman- α applied to recoil energy and momentum distribution measurements of H and D atoms in dissociation processes. - Depending on the objective, different experimental conditions and systems have been employed, for instance atomic beam configuration, mass-spectroscopic ion detection, field ionization in cryogenetic environment, and for Doppler measurements laser induced fluorescence.

Generation of Coherent Radiation Below
100nm in Hg Vapor

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We have studied two-photon resonant third harmonic generation of coherent radiation below 100nm in Hg. Output near 93 nm was produced with input radiation from a frequency doubled dye laser tuned near the $6s^2-6s6d$ two-photon resonance in Hg at 280.3nm, and output near 83nm was produced with radiation from a tunable KrF excimer laser tuned near the $6s^2-6s10s$ two-photon resonance in Hg at 248.8nm.

Several features of the observed output are noteworthy: 1) At high vapor density and input intensity, saturation accompanied by two-photon absorption was observed leading to the peak output being observed for tunings slightly off exact two-photon resonance. This result has been predicted theoretically¹; 2) Resonant enhancement of the 93nm generation was observed on both the $6s6d\ ^1D_2$ and $6s6d\ ^3D_2$ states, with a deep minimum between the two enhancements; 3) An additional broad enhancement of the 93 nm generation was observed at a wavelength which coincides with the dipole forbidden $6s^2\ ^1S_0 \rightarrow 6s7p\ ^1P_1^o$ two-photon resonance.

The generation of tunable coherent radiation in the wavelength region below 100nm opens up a new region of the spectrum to study by high resolution spectroscopy. Of particular interest will be the generation of broadly tunable radiation by 4-wave mixing

in which one laser is tuned to a two-photon resonance and a second tunable laser is added to produce widely tunable, narrow band output in the vacuum ultraviolet between 83 and 120nm.

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WA3-1

Spectroscopy of CO and NO Using Coherently Generated VUV

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NON-LINEAR MIXING II

Generation of Narrowband Tunable VUV Radiation

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Nonresonant sum- and difference-frequency mixing of the fundamental (ω_L) and the second harmonic (ω_{uv}) output of a powerful narrowband pulsed dye laser excited by a Nd-YAG laser has been investigated in Xe and Kr. The sum-frequency $\omega_{vuv} = 2\omega_{uv} + \omega_L$ is tunable in spectral regions of negative dispersion between 110 nm, and 130 nm. The maximum VUV pulse power exceeds 20 W ($5 \cdot 10^{10}$ photons/pulse). The difference frequency $\omega_{vuv} = 2\omega_{uv} - \omega_L$ provides VUV light pulses with up to 60 W ($2.3 \cdot 10^{11}$ photons/pulse) at wavelengths between 185 nm and 207 nm. Coherent VUV light of shorter wavelength (159.5 nm to 186.6 nm) is obtained by mixing the UV dye laser radiation with the infrared output (ω_{IR}) of the Nd-YAG laser ($\omega_{vuv} = 2\omega_{uv} - \omega_{IR}$). With UV light of shorter wavelength ($\omega'_{uv} = \omega_{uv} + \omega_{IR}$) the difference frequency conversions $\omega_{vuv} = 2\omega'_{uv} - \omega_L$ and $\omega_{vuv} = 2\omega'_{uv} - \omega_{IR}$ allow the generation of VUV light with wavelengths between 122.6 nm and 160 nm. Thus with a single dye laser which is operated in the most efficient operating range of Nd-YAG laser pumped dye lasers ($\omega_L = 550 - 670$ nm) the investigated conversion schemes generate intense coherent VUV light which is continuously tunable between 110 nm and 210 nm. Besides the nonresonant frequency conversion two-photon resonant frequency mixing has been studied in detail in Xe and Hg. In these experiments the resonant enhancement of the nonlinear processes increased the VUV pulse power to values in the range of 0.5 to 10 KW.

In Xe the two-photon transitions $5p - 6p$, $5p - 7p$ and $5p - 4f$ have been used for a resonantly enhanced generation of VUV light at $\omega_{vuv} = 2\omega_1 \pm \omega_L$, where ω_1 is the two-photon transition frequency and ω_L is tunable in the spectral range of $\lambda_L = 220 - 760$ nm. While the sum-frequency generates radiation in the XUV ($\lambda_{vuv} = 73$ nm to 101 nm) the difference frequency is continuously tunable between 129 nm and 220 nm. The experimental results provide, for example, detailed information

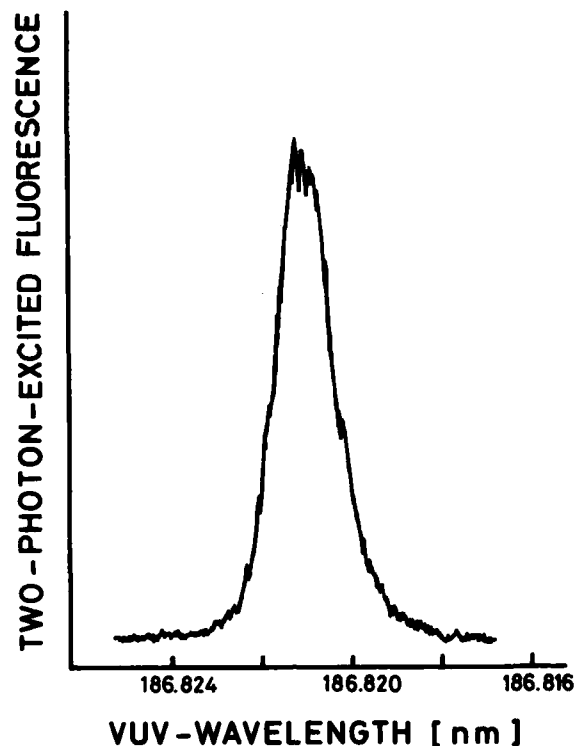


Fig. 1 Two-photon excitation of the $4p[1/2]0$ level of Argon. The narrowband VUV radiation (with a pulse power of more than 2KW) is generated by difference-frequency mixing in Hg vapor. The excitation is observed via the transition ($4p[1/2]0 \rightarrow 4s[3/2]1$) at $\lambda = 751.5$ nm

on conversion limiting saturation phenomena and their dependence on the input power and the detuning of ω_1 from the two-photon resonant value.

In Hg the resonant frequency conversion is investigated for the resonances $6^1S_0 - 7^1S_0$ and $6^1S_0 - 6^1D_2$. Below the ionization limit intense VUV is observed at the sum-frequency in the wavelength range of $\lambda_{\text{vuv}} = 119 \text{ nm} - 127 \text{ nm}$ in the vicinity of excited states $6s \text{ np } (n \geq 9)$. Continuously tunable radiation is provided by the sum-frequency above the ionization limit ($\lambda_{\text{vuv}} = 111.7 - 116 \text{ nm}$) and by the difference frequency at $\lambda_{\text{vuv}} = 177 - 188 \text{ nm}$.

While the intensity of the VUV light generated by the nonresonant frequency conversion is sufficient for precise linear spectroscopy, the high pulse power obtained by the resonant frequency mixing allows the application for nonlinear spectroscopy in the XUV (see Fig. 1). Since very intense narrowband VUV is generated at wavelengths as short as 120 nm the two-photon excitation will be advantageous for doppler-free investigations in the XUV at $\lambda = 60$ to 100 nm.

Generation of Tunable, Coherent 79 nm Radiation by Frequency Mixing

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SUMMARY

In this paper, we report on the generation of tunable radiation around 79 nm, using frequency mixing of two ArF* photons and a visible dye laser photon. In contrast to harmonic generation with a rather limited tuning range, this method combines the potential of high power excimer lasers (up to 30 W peak powers have recently been measured¹ at the third harmonic of ArF*) with the broad tunability of dye lasers. The ArF* lasers used in the present experiments is a modified version of a system described previously.² The output characteristics of the ArF* laser are energy 200 mJ, bandwidth 0.5 cm^{-1} , pulse duration 7 ns, and wavelength 193.55 nm. The tunable dye laser was operated with stilbene 420, tunable from 416 nm to 458 nm, with an output energy of 5 mJ, bandwidth 0.3 cm^{-1} , and pulse duration 15 ns. With these lasers, up to 200 mW peak power of mixed radiation have been measured with hydrogen as nonlinear medium. This radiation has a bandwidth of less than 0.8 cm^{-1} , if gaussian lineshapes are assumed. In order to demonstrate the usefulness of the system as a spectroscopic tool, the source was tuned across the $^2P_{1/2}$ 9d' autoionizing resonance in argon. Fig. 1 shows the measured absorption profile normalized to the data from Hudson and Carter³. The bandwidth of the source can be tested by scanning across the $^2P_{3/2}$ 13d states which have an estimated doppler width of 0.22 cm^{-1} .

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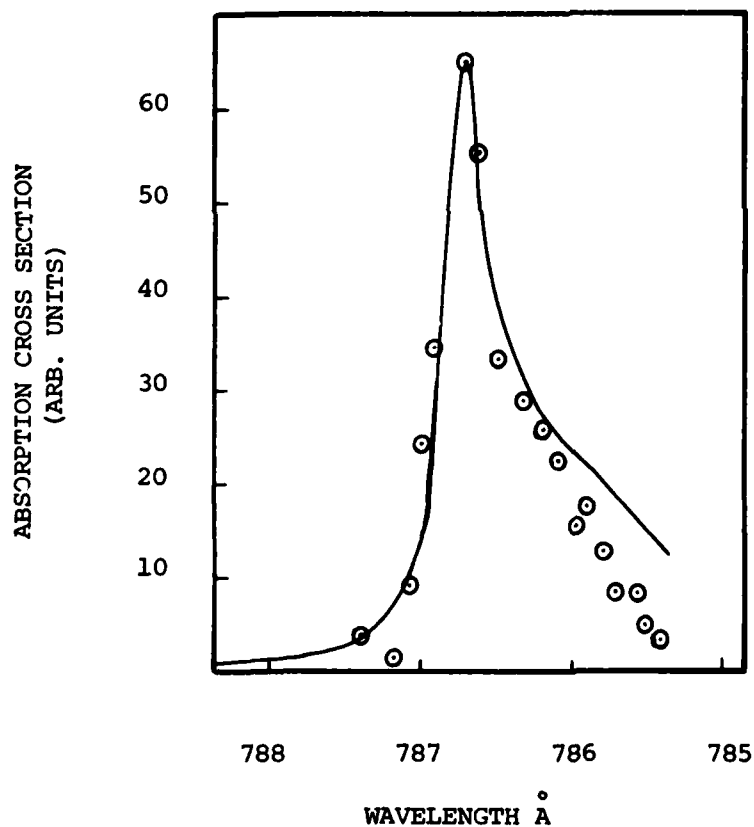


Fig 1.: Measured absorption profile $^2P_{1/2}$ $9d'$ auto-ionizing resonance of argon, normalized to data from Hudson and Carter³.

Upconversion of Laser Radiation
to γ -Ray Energies

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Reviewed here are studies of the general problem of multiphoton processes which might occur at the nuclear level when one of the photons belongs to an intense radiation field from a laser at optical frequencies. It has been predicted that sufficient laser intensities could stimulate a component step involving the nuclear radiations. Transition probabilities have been found which relate the probabilities for γ -ray transitions induced by the optical radiation to the Breit-Wigner cross sections for the absorption or emission of single γ -photons. The most important results have concerned the isolation of the multiphoton Mössbauer channel which preserves the natural linewidth of the process in solid media. This channel was found to have a relative weight comparable to the conventional Debye-Waller factor for the transition. In emission the induced processes of both Anti-Stokes Raman scattering and two-photon emission have been considered and both are found to lead to the generation of tunable γ -radiation. Transition probabilities are calculated to be large enough to support the production of usable spectral sources that could be tuned over a range of γ -ray energies approximately a thousand times greater than can be realized by conventional Mössbauer techniques. Analogous possibilities for the detection and tuning of nuclear reactions induced by intense optical fields also have been considered in this review.

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